Report on MASTS DSF Small Grant project:

A new approach for measuring the sediment tracer thorium-234 in deep sea samples

Scottish Association for Marine Science (SAMS), Oban, UK

Robert Turnewitsch, Jirina Stehlikova

1. Background

The spatiotemporal distribution of the quantity and quality of depositing sediments controls benthic food supply and biodiversity, and detailed knowledge of geographical distributions of sediment dynamics facilitates exploration of economically relevant iron-manganese (Fe-Mn) deposits in the deep sea. Consequently, an in-depth mechanistic insight into the processes involved in the formation and dynamics of sedimentary deposits in the deep sea is essential. One way of learning more about the mechanisms that control sediment dynamics near the sediment/water interface is the measurement of the naturally occurring particle tracer thorium-234 (^{234}Th). Measurements of ^{234}Th in water samples are comparatively straightforward. However, because of much lower specific radioactivities, ^{234}Th measurements in sediments are more difficult. This Small Grant project aimed to contribute to the development of a new approach that we hope will facilitate the non-destructive measurement of ^{234}Th in comparatively small amounts of sediment, with the added advantage of being able to use the sediment for further analyses after completion of the ^{234}Th analyses. As one of the steps in the method development, the counting efficiency of the sediment pellets that were used to measure the radioactivity in a low-level betacounter had to be quantified. To quantify the counting efficiency that results from self-absorption of radiation within the sample and the specifics of the counter geometry, a ^{234}Th standard was required. The requested MASTS DSF funds were used to purchase two ampoules of a certified ^{238}U standard solution (IRMM-056) that is in radioactive equilibrium with ^{234}Th and can be used as a ^{234}Th standard.

2. Laboratory work

The method development was carried out with sediments that were sampled from the Tonga Trench in the Southwest Pacific. At the time of this experiment, the sediments were old enough to allow for the assumption that there is no natural excess ^{234}Th anymore (excess ^{234}Th is the ^{234}Th fraction that is due to sediment input from the overlying water column rather than ^{238}U decay within the sediment) and that the natural beta-radioactivity of the sample material (the 'background') is stable on the characteristic time scale of ^{234}Th (several months) and on shorter time scales. Sediments were dried and ground. Sediment pellets of reproducible geometry and a mass of between ~2.0g and ~2.4g were then made. During pellet production, known amounts of ^{238}U standard solution (1500µl which equates to approximately 1.74g of solution) were mixed into the sediment, ensuring that the solution is well homogenised within the sediment. The homogenisation was monitored by addition of a dye to the sediment. All handling of the sediments and standard solution was carried out in a fume cupboard in a dedicated radioisotope laboratory. In total, 13 ^{238}U-spiked sediment pellets were made: 5 pellets contained sediments from the deep Tonga-Trench axis (~10810m water depth) and 8 pellets contained sediment from the oceanward trench rim (~6255m water depth) (Fig. 1).

All pellets were measured directly and non-destructively in a Risø Low-Level Beta GM-25-5 Multicounter (the multicounter contains 5 detectors so that 5 samples can be measured simultaneously). The pellets sat in circular nylon dishes and were covered by a Mylar foil that was stuck to a thicker transparent plastic film to prevent any alpha particles from contributing to the detection signal (Fig. 1). Each pellet was measured several times over more than 6 ^{234}Th half-lives to ensure detection of potential artificial ^{234}Th/^{238}U disequilibria that could have resulted from inhomogeneous distribution of non-particle-reactive ^{238}U vs. particle-reactive ^{234}Th in the pellets. Each of the 13 standard pellets was measured in each of the five detectors at least three times. For each pellet, the measured activities from all five detectors were averaged.

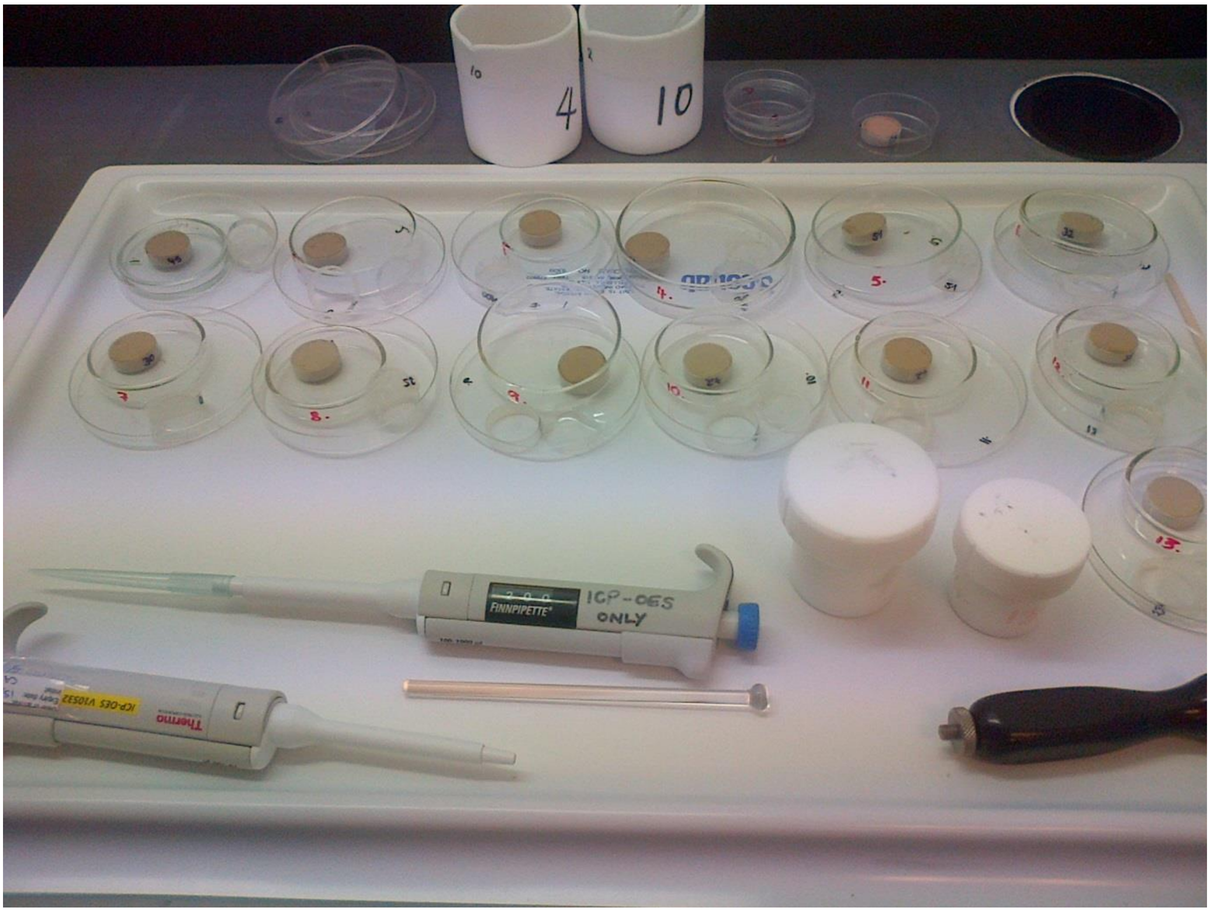

Fig. 1. The 13 sediment pellets that were spiked with the certified ^{238}U standard solution (IRMM-056) that is in radioactive equilibrium with ^{234}Th. The pellets sat in circular nylon dishes and were covered by a Mylar foil that was stuck to a thicker transparent plastic film to prevent any alpha particles from contributing to the detection signal.

3. Results, discussion and application

The counting efficiencies for the 13 pellets ranged between 7.711% (or 7.711 counts registered by the counter per 100 true ^{234}Th disintegrations occurring in the whole sediment pellet) and 9.706% (or 9.706 counts registered by the counter per 100 true ^{234}Th disintegrations occurring in the whole sediment pellet). There is some evidence to suggest that efficiency values for samples collected from the trench-axis sites (stations C1 and C2) are somewhat lower than efficiency values for samples collected from the trench-rim sites (stations C4 and C6). Counting efficiencies have been plotted against 'background' radioactivity (the natural beta-radioactivity after decay of all excess ^{234}Th and before addition of the standard solution) per pellet (Fig. 2), specific background activity (i.e., background activity normalised by the mass of the pellet (Fig. 3), dry bulk density of the pelletised sediment (Fig. 4), and absolute pellet mass (Fig. 5).

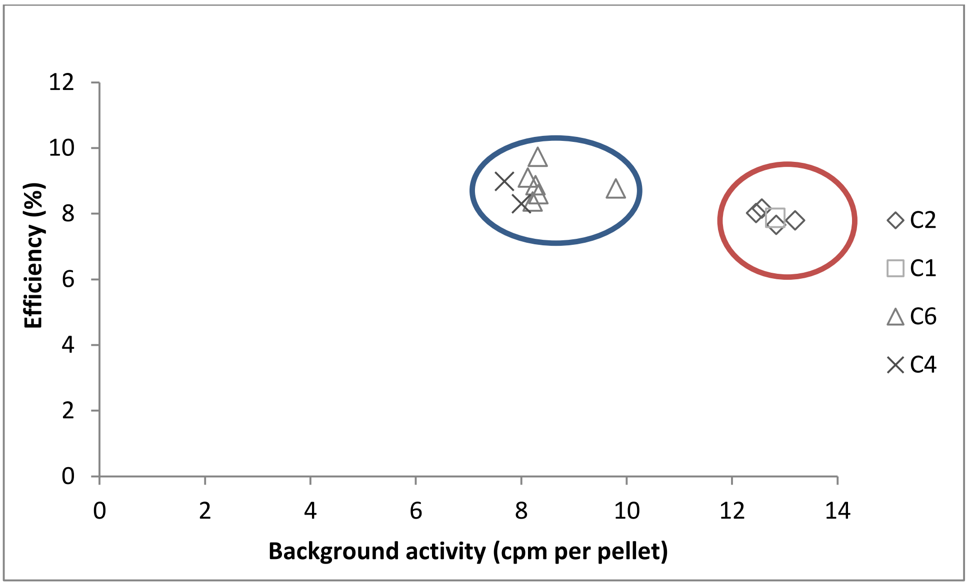

Fig. 2. Counting efficiencies plotted versus background activity of the total sediment pellet (cpm: counts per minute). Trench-axis sites (red ellipse): C1 and C2; trench-rim sites (blue ellipse): C4 and C6.

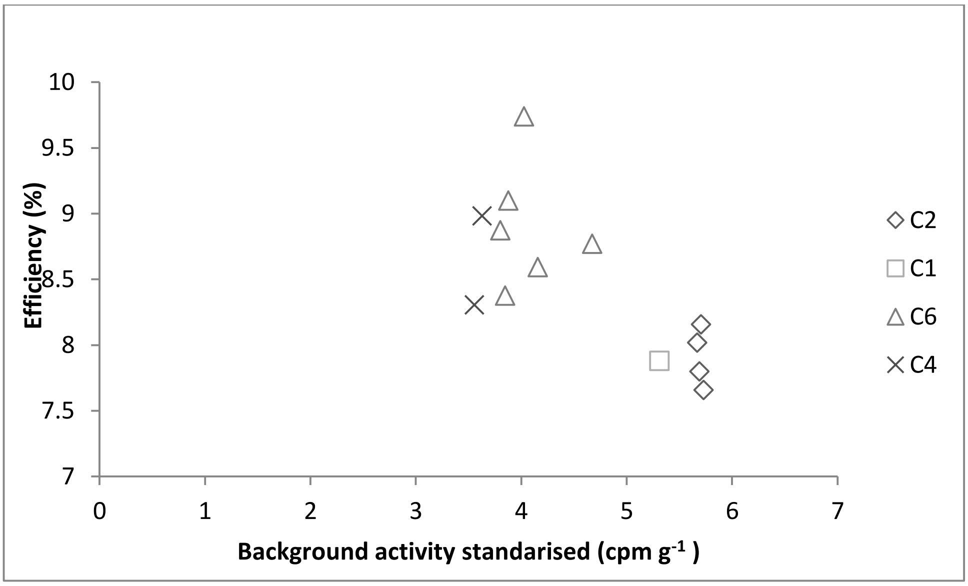

Fig. 3. Same as Fig. 2, but plotted versus mass-normalised background activity.

Interestingly, there was no influence of dry bulk density, and only a weak influence of absolute pellet mass, on counting efficiency, at least over the dry-bulk-density and mass ranges encountered in this study (Fig. 4 and 5). By contrast, the relationships between natural background activity and mass-normalised background activity on the one hand and counting efficiency on the other hand are somewhat clearer (Fig. 2 and 3). Further experiments are required to find out whether there is in fact a systematic influence of (the relative magnitude) of background activities on calculated counting efficiencies or if there are other factors that have not been considered so far and only cause an apparent link between backgrounds and counting efficiencies.

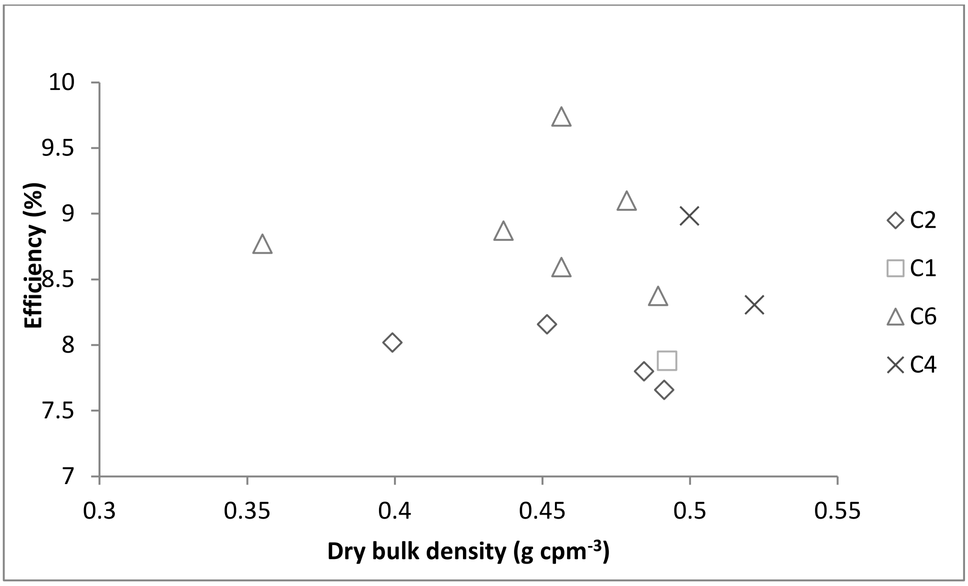

Fig. 4. Same as Fig. 2, but plotted versus dry bulk density of the pelletised sediment.

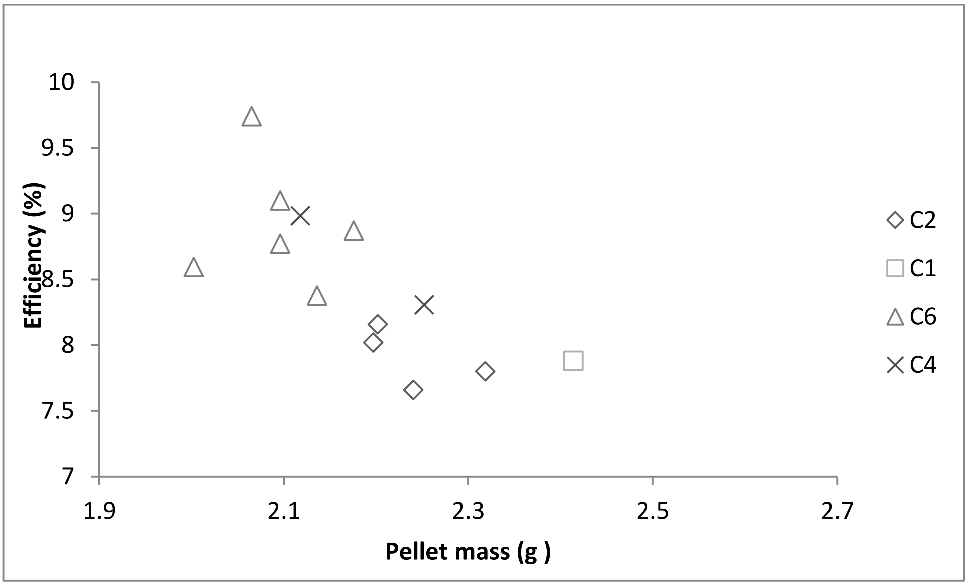

Fig. 5. Same as Fig. 2, but plotted versus absolute pellet mass.

Despite such open methodological questions, the calculated counting efficiencies are sufficiently robust (their scatter is sufficiently small) to apply them to un-spiked sediment samples. An example of a sedimentary excess ^{234}Th profile from the Tonga-Trench axis that was derived by applying the newly calculated counting efficiencies to the results from un-spiked sediments is shown in Figure 6. It turns out that the topmost centimetres of the trench-axis sediments contain detectable amounts of ^{234}Th, with the activities approaching zero in sediments deeper than 3cm. The absolute values are fairly typical for surface sediments in the deep sea and indicate that there are active sediment dynamics taking place on time scales shorter than a couple months. Historically, on such short time scales, hadal trenches have been viewed as very un-dynamic and quiescent environments. The presence of detectable amounts of ^{234}Th in surface sediments indicates that this picture may not be universally true. This preliminary finding has potential implications for our understanding of food supply to hadal trenches and also the speed with which early-diagenetic biogeochemical processes are running in hadal sediments.

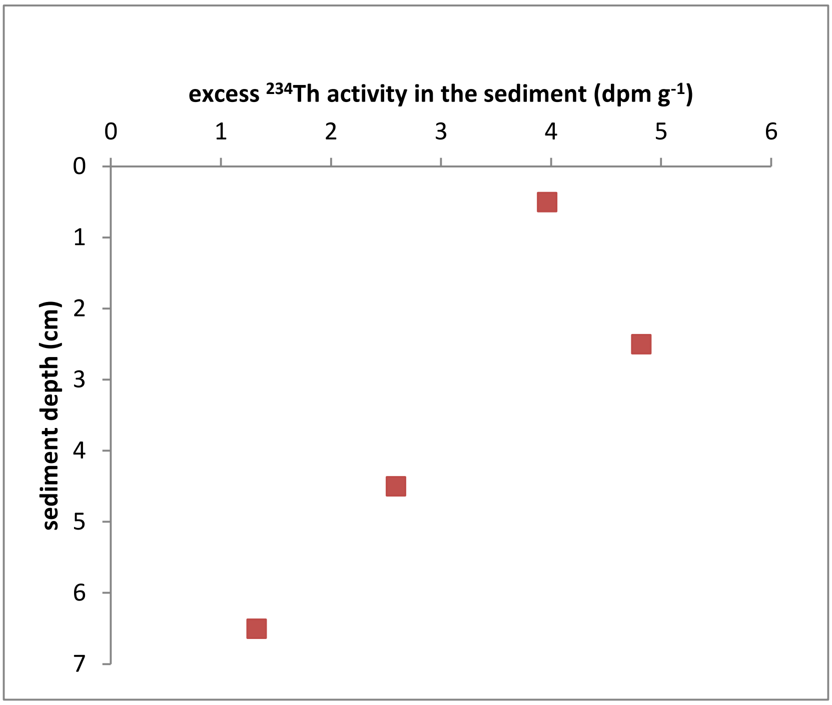

Fig. 6. Sedimentary profile of excess ^{234}Th for station C2 in the Tonga-Trench axis. dpm: disintegrations per minute.

Finally, in terms of the methodology, here, the decision of which efficiency value to use for which un-spiked sample depended upon the geographical location of the sample, mass of the pellet, background activity, sediment depth, and the detector used for measurement. In future applications this close alignment of factors with properties of un-spiked samples may not always be possible. But the relatively low extent of variability of counting efficiencies between deep-sea samples from different localities and sediment depths is very encouraging, and the values from this study may well turn out to be fairly representative for non-carbonaceous deep-sea deposits in general.