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Actinides, accelerators and erosion

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Abstract. Fallout isotopes can be used as artificial tracers of soil erosion and sediment accumulation. The most commonly used isotope to date has been ^{137}Cs. Concentrations of ^{137}Cs are, however, significantly lower in the Southern Hemisphere, and furthermore have now declined to 35% of original values due to radioactive decay. As a consequence the future utility of ^{137}Cs is limited in Australia, with many erosion applications becoming untenable within the next 20 years, and there is a need to replace it with another tracer. Plutonium could fill this role, and has the advantages that there were six times as many atoms of Pu as of ^{137}Cs in fallout, and any loss to decay has been negligible due to the long half-lives of the plutonium isotopes. Uranium-236 is another long-lived fallout isotope with significant potential for exploitation as a tracer of soil and sediment movement. Uranium is expected to be more mobile in soils than plutonium (or caesium), and hence the ^{236}U/Pu ratio will vary with soil depth, and so could provide an independent measure of the amount of soil loss. In this paper we discuss accelerator based ultra-sensitive measurements of plutonium and ^{236}U isotopes and their advantages over ^{137}Cs as tracers of soil erosion and sediment movement.

1 Introduction

Soil erosion in Australia is recognized as a major ongoing issue, and its mitigation by sustainable land management practices will be an ongoing process into the future. The effects of climate, for instance, will directly impact soil stability, and in traditional agricultural areas will need to be managed alongside the increasing pressure to provide enough food for our expanding population. The need to produce more food will require changes in farming operations and is also likely to force expansion of agriculture into areas not currently used. It is likely that this will be into areas with soils of poorer structure and less resistance to erosion. The ability to estimate decadal-scale erosion losses in association with these changes in land management practice will therefore be important in the coming years.

Radio-isotopic tracers, particularly fallout ^{137}Cs, have been used for many years to study soil and sediment movement, and in conjunction with modelling, provide a means to assess the effectiveness of individual management practices. Assessing the effects of land management on soil erosion is however a difficult process, because of the spatial and time scales involved. Significant reliance is therefore placed on modelling, and isotopic tracers provide a valuable tool for testing and developing such models.

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The total ^{137}Cs activity dispersed by the atmospheric nuclear weapons tests of the 1950's and 1960's was of the order of 1000 PBq [1]. This, and the ability to determine the ^{137}Cs concentration readily by counting the 662 keV γ-ray emitted when the ^{137}Cs ($t_{1/2}$ = 30a) decays, have allowed significant use of ^{137}Cs as a tracer of soil and sediment transport that has occurred in the time since fallout deposition. In the Southern Hemisphere however, fallout levels were significantly lower compared to those in the north, and combined with the steady decline in ^{137}Cs activity as a result of radioactive decay, is beginning to limit the analytical uncertainties of the measurements. As a consequence, many ^{137}Cs erosion applications will become untenable within the next ~20 years. This will constrain the future utility of fallout Cs as a tracer, and there is a real need for an equivalent replacement.

In addition to ^{137}Cs, the plutonium isotopes $^{239, 240}$Pu were also distributed around the globe as a result of the nuclear weapons tests. The total released activity was an order of magnitude less than that of Cs, and plutonium fallout from the tests was not widely monitored; however the historical fallout pattern is believed to show a structure similar to that of caesium [2]. In terms of its suitability as a replacement for ^{137}Cs, there is growing evidence that Pu and Cs display similar particle-reactive behaviour in terrestrial environments [3-5]. Furthermore, the long half-lives of ^{239}Pu ($t_{1/2}$ = 24,110a) and ^{240}Pu ($t_{1/2}$ = 6561a) has resulted in the radioactive decay of only

0.2% of the original fallout, and permit their use as monitors of net soil redistribution for many decades into the future.

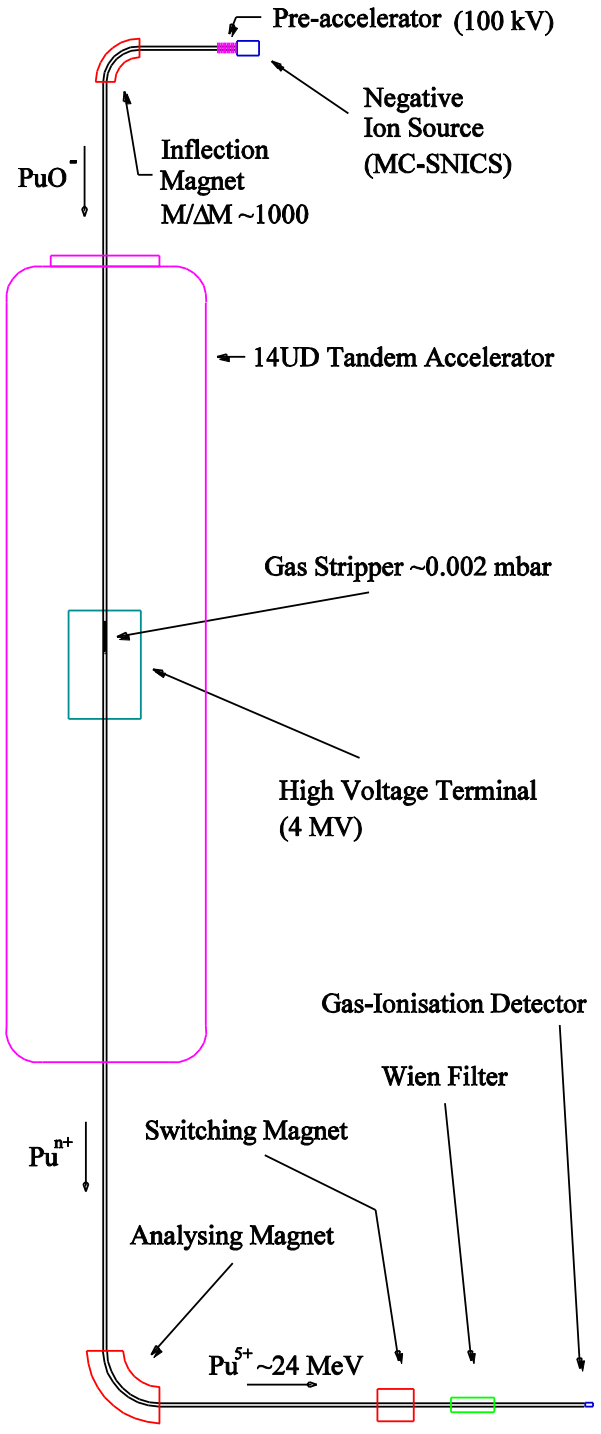

Fig. 1. Essential features of the ANU AMS system for the measurement of Pu.

The technique of Accelerator Mass Spectrometry (AMS) counts atoms directly, rather than measuring their radioactive decay. This is of note, because the nuclear tests yielded over six times as many atoms of $^{239+240}$Pu as ^{137}Cs atoms. AMS plutonium measurements also confer a number of advantages over caesium measurements [6], including reduced counting times, the near absence of background interference and improved statistical precision. A further significant advantage is that the ^{240}Pu/^{239}Pu ratio from local nuclear weapons test sites can differ from the global average. This can permit assessment of the significance of any "local" contribution to the inventory [7]. This information cannot be gained from ^{137}Cs measurements alone. In addition, the AMS technique also offers the opportunity to investigate a new, complementary tracer to plutonium for the assessment of soil loss and movement: fallout ^{236}U.

2 Plutonium measurements

Figure 1 shows a schematic representation of the AMS system at the Australian National University (ANU) as configured for Pu measurements. Soil and sediment samples are prepared at the ANU for AMS analysis based on the techniques described in [4]. This entails addition of a ^{242}Pu spike to the homogenised soil or sediment material, and the plutonium is then leached from the sample with hot nitric acid and purified using ion exchange columns. The extracted material is then dispersed in an iron-oxide matrix and pressed into AMS sample holders.

Plutonium isotopes are currently measured with the AMS system using a "slow-cycling" technique, wherein the isotopes ^{242}Pu, ^{240}Pu and ^{239}Pu are injected as PuO^- ions into the 14UD accelerator in turn by adjusting the magnetic field in the mass-analysing (Inflection) magnet (figure 1). Details of the AMS system and measurement methodology relevant to actinide measurements are given in [6, 8]. The ^{242}Pu, ^{240}Pu and ^{239}Pu isotopes are normally measured for 1, 3 and 2 minutes, respectively, with the sequence being repeated as many times as is necessary, with three loops being typical. The isotopic ratios ^{239}Pu/^{242}Pu, ^{240}Pu/^{242}Pu and ^{240}Pu/^{239}Pu are determined from the data, and ^{239}Pu and ^{240}Pu concentrations in the original sample material deduced from the known amount of added spike. A typical ^{239}Pu spectrum recorded with the ANU AMS system is shown in figure 2.

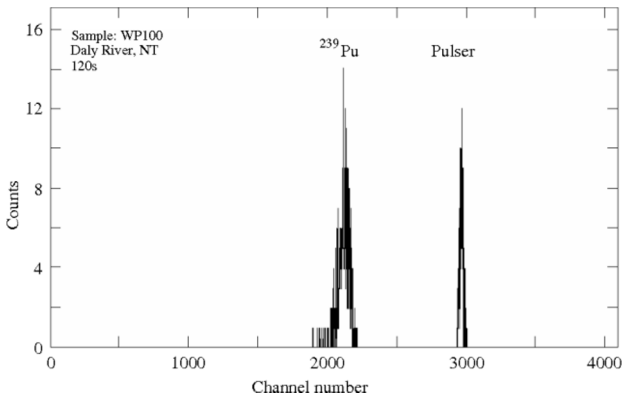

Fig. 2. Typical ^{239}Pu spectrum obtained from a soil sample. The ~20 g soil sample was collect from the top 0–5 cm of the soil profile and yielded a ^{239}Pu peak of over 600 counts in a 2 minute collection period.

In soils the concentrations of fallout isotopes vary with depth as a consequence of bioturbation, mechanical

processes that move soil grains down the profile, and also possibly as a result of movement in solution. The shape of the depth profile is determined by factors such as soil type, organic matter content, porosity and by the chemical properties of the isotopic tracer (i.e. by how well the tracer binds to the soil particles). Caesium-137 and the plutonium isotopes bind tightly to soil particles, and there is good evidence that the soil depth profiles of both elements peak at or near the surface, at approximately the same depths (figure 3) [5]. There is also evidence that their concentrations can be well correlated in soils and sediments collected from different land-use types [e.g. 4, 9]. The similar behaviour of Pu and Cs in soils, combined with the superior statistical precision of the AMS measurements, should permit Pu measurements to be referenced against existing ^{137}Cs soil inventory data. Quantification of the effects of recent land use change, through such referenced data sets, could provide a means to assess changes in soil redistribution associated with modern changes in land management practices, and will also provide new data sets with which to test and validate soil and sediment re-distribution models.

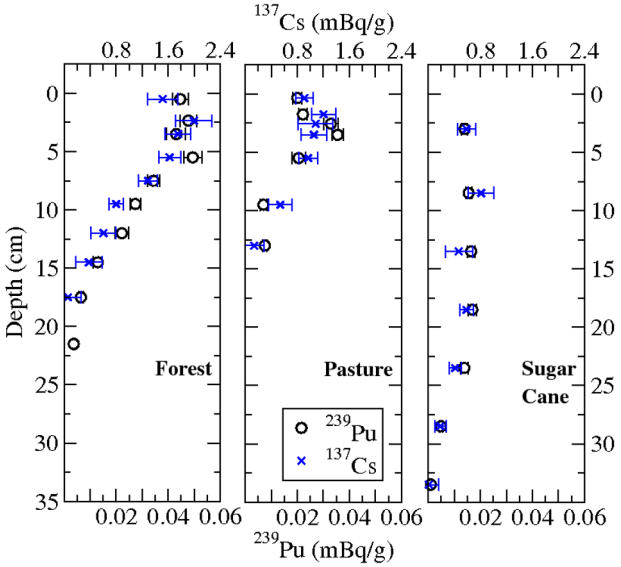

Fig. 3. Depth profiles for ^{137}Cs and ^{239}Pu in soils collected from different land-use types. From [5].

3 Uranium-236

Comparison of ^{137}Cs depth profiles from eroded sites with those from undisturbed reference sites has long been used to deduce the depth of soil material that has been lost to erosion. This technique assumes either that the soil characteristics at the reference site match those at the site under investigation, or that any differences in the characteristics are not significant factors in the loss of soil. The use of Pu isotopes in place of ^{137}Cs is subject to the same assumptions.

Bomb-produced ^{236}U also has a long half-life ($t_{1/2}$ = 23.42 Ma), and potential as a complementary tracer to plutonium for soil loss and sediment movement studies. Uranium is expected to be more mobile in soils than plutonium, particularly in acidic soils [10]. This difference is likely to give rise to differently shaped depth profiles – ^{236}U concentration depth profiles should peak at a greater depth than those for plutonium. This may get around a limitation of using Pu alone, i.e. that the loss of only a thin layer of surface soil can result in the loss of much of the Pu, which is concentrated in the top few cm of the soil. Determination of the actual amount of soil loss is then sensitive to the assumed depth profile of Pu close to the surface, and is a significant source of uncertainty. If the ^{236}U/Pu ratio varies with depth, the ratio in the surface soil at an eroded site could provide a semi-independent measure of the amount of soil loss. The ratio could also provide a means to check how well the sampling and reference site soil characteristics match. Furthermore, the measurement of this ratio in transported sediment could also provide valuable information on the average depth from which sediment has been derived by the combination of surface wash and gullying processes.

AMS is by far the most sensitive technique with which to measure fallout ^{236}U in environmental samples, and the only technique currently capable of measuring ^{236}U at the requisite sensitivity [11]. Furthermore, extraction and measurement of ^{236}U and the Pu isotopes from the same environmental material has been demonstrated at the levels necessary [12, 13]. There is, however, very little data in the literature regarding concentrations of fallout ^{236}U in soils, and even less regarding the use of ^{236}U as an environmental tracer, although first attempts have been reported recently [14, 15].

4 Summary

In an era where Australian agriculture is undergoing changes in traditional management practices, and expansion into new areas likely to be more susceptible to erosion, AMS actinide measurements can provide a long-term solution to the declining sensitivity of ^{137}Cs measurements used for the assessment of soil loss and sediment movement. The superior precision of the AMS plutonium measurements, compared to ^{137}Cs gamma-spectroscopy data, and the opportunity for new methods for erosion analysis using the new fallout isotope ^{236}U, will allow for continued improvement and validation of soil and sediment re-distribution models.

It is noteworthy that the ^{240}Pu/^{239}Pu ratio, routinely determined by the AMS analysis, can indicate the significance of any contribution to the fallout inventory arising from "local" nuclear weapons tests, a factor that cannot be taken into account from ^{137}Cs measurements alone. This could be important in the Australian context, as candidate areas for the expansion of agriculture potentially fall within regions influenced by fallout from nuclear weapons tests carried out in Australia.

There is also significant potential to make use of the extensive database of ^{137}Cs results that already exist, via data sets referenced to AMS plutonium measurements. Such data could be used to assess the efficacy of changes in land management methods focused on minimising soil loss.

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