A Letter Of Intent To
The ISOLDE and Neutron Time-Of-Flight Experiments Committee (INTC)

PREPARATION & IMPLANTATION OF CARRIER-FREE
\(^{115m}\)Cd IN CALCIUM SALT

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1 INTRODUCTION

The use of radioisotopes has significantly assisted study of the behaviour of nutrients and heavy metals in soil. Many heavy metals have useful radioisotopes that can be used to study retention and plant uptake processes in soil, including \(^{109}\)Cd and \(^{115m}\)Cd, \(^{60}\)Co, \(^{64}\)Cu, \(^{202}\)Hg, \(^{54}\)Mn, \(^{63}\)Ni, \(^{210}\)Pb, \(^{73}\)As and \(^{65}\)Zn. The isotopic dilution technique allows partitioning of metals into isotopically exchangeable (labile) and non-exchangeable (fixed) pools. The labile pool is considered to reflect the chemical reactivity and/or potential bioavailability of metals in soils [1-3].

When an isotope is introduced into a soil suspension, its distribution will reflect that of the corresponding labile metal pool in the soil. Therefore, a quantity of an isotope added to a system will distribute itself between the solution and the exchangeable phases. Hence, the total concentration of radio-labile metal in the soil \([M_E]\) (mg kg\(^{-1}\)) is calculated as:

\[
[M_E] = [M_{Soil}] \left( k_d^* + \frac{V}{W} \right) \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (i)
\]

where \([M_{Soil}]\) is the concentration of metal in solution (mg L\(^{-1}\)), \(k_d^*\) is the isotopic distribution coefficient (L kg\(^{-1}\)), and \(V\) and \(W\) are the volume of solution (L) and the mass of soil (kg),

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respectively [1]. Isotopic dilution techniques can be used to investigate ageing or ‘fixation’ of metals in soils, that is the slow transformation of metals from labile to non-labile pools [1,4].

The advantage of using the method of isotopic dilution is that it does not disturb the existing chemical equilibria in the system and it provides accurate quantification of metal lability and fixation processes at environmentally realistic metal concentrations. However, when metals are present at low concentrations, the presence of sub-micron colloidal particles holding non-labile metal can cause large errors in the determination of labile metal. These colloidal particles may be left suspended in solution even when centrifugation or micro-filtration is used for the phase separation operation during the measurement of labile metal in soils and sediments.

2 MOTIVATION AND APPROACH

The need to quantify this phenomenon arises for two reasons. Firstly, the incidental assay of non-labile metal associated with sub-micron colloids as part of \([M_{Solv}]\) in equation (i) represents a significant error in the determination of labile soil metal \([M_E]\). The determination of radio-labile metal is rapidly becoming one of the most popular geochemical tools for following trace metal dynamics. Secondly determining the conditions under which non-reactive trace metals within sub-micron colloids occur has significant implications for geochemical transport models.

Briefly, a double-labelling technique will be employed to quantify the extent of colloidal interferences in the determination of Cd lability in selected soils. This can be achieved by the combined use of \(^{109}\)Cd and \(^{115m}\)Cd whose max \(\gamma\)-energy lines do not overlap \((E_{\gamma,\text{Max}} ^{^{109}\text{Cd}} = 88 \text{ keV}; E_{\gamma,\text{Max}} ^{^{115m}\text{Cd}} = 1290 \text{ keV})\). Selected soils, which are essentially Cd-free, will be spiked with natural Cd and \(^{109}\)Cd, simultaneously while in electrolyte suspension. This will induce some fixation of \(^{109}\)Cd-labelled Cd within geo-colloidal structures following an equilibration time of several months. To measure the concentration of labile Cd, soil suspensions will then be spiked with \(^{115m}\)Cd. Determining the isotopic ratio \(^{109}\text{Cd}/^{115m}\text{Cd}\) would then provide an estimate of \(C_{dE}\) (adapting equation (i)). However, such an assay is subject to errors arising from fixed \(^{109}\)Cd in suspended sub-micron particulates. Therefore, a sample of filtered suspension will then be extracted with Chelex 100 (a cation exchange resin) for 1-2 h. The resin will effectively ‘sample’ Cd only from the labile pool and so radio-assay of the resin will provide the true isotopic ratio in the radio-labile pool. Effectively,
comparison of the isotopic ratios in the whole solution and resin phases provides a means of very accurately quantifying the extent of formation non-labile colloidal Cd.

\(^{115m}\)Cd is essential for conducting this novel experiment. Although, the production of a carrier-free \(^{115m}\)Cd can be challenging we believe it is a unique opportunity to produce it at ISOLDE, which is the best facility for this purpose in the world.

3 EXPERIMENTAL OVERVIEW

\(^{115m}\)Cd ion beams will be implanted into a suitable catcher, which is in our case either CaCl\(_2\) or Ca(NO\(_3\))\(_2\). Calcium salt will be compressed to obtain multiple pills with a total mass of about 0.3 g. After transport to the University of Nottingham, the implanted \(^{115m}\)Cd will be brought into a neutral aqueous solution by dissolving the calcium salt in MILLIQ water. This procedure is favoured over the use of metallic foil because our sorption experiment requires the presence of \(^{115m}\)Cd\(^{2+}\) in a solution of ~ 0.01 M Ca\(^{2+}\) (as CaCl\(_2\) or Ca(NO\(_3\))\(_2\)) at neutral pH. Small amounts of pure H\(_2\)O\(_2\) (or pure HNO\(_3\) as oxidizing agent) will be added to the solution to ensure that all the \(^{115m}\)Cd is present as \(^{115m}\)Cd\(^{2+}\) rather than any other oxidation state.

Radioactive Cd-isotopes are produced at ISOLDE by proton-induced fission using UC\(_x\) target and ionized RILIS (resonance ionization laser ion source). The ion yield of \(^{115}\)Cd obtained in a recent study using UC\(_x\)/graphite and RILIS was 1.1 \(\times\) 10\(^8\) ions/\(\mu\)C [5]. Using the combination of fission and resonant laser ionisation (which favours the high spin isomer), the ratio of \(^{115m}\)Cd/\(^{115}\)Cd will be high and we expect to collect about 6 MBq of \(^{115m}\)Cd in 6 shifts. The University of Nottingham has a handling capacity of 1.0 GBq. Our lab has a handling permit for up to 37 MBq of \(^{115m}\)Cd.

4 WHY ISOLDE?

The required facility for the production of \(^{115m}\)Cd must have the key elements: efficiency, high yield, selectivity and experience of the collaborating team. Researchers at ISOLDE have implanted samples with Cd isotopes for solid-state studies and spectroscopy. ISOLDE uses RILIS technique for the separation of isotopes, which is a very efficient and selective online separation technique. Therefore, the only place at present, which can produce a carrier-free
\(^{115m}\text{Cd}\) is CERN-ISOLDE. For these reasons and the uniqueness of ISOLDE facility, it is the most suitable facility for the production of \(^{115m}\text{Cd}\).

5 BEAM TIME REQUEST

We ask for a total of 6 parasitic shifts since the collection can be performed at the GLM\(^4\) beam line in parallel to any main experiment (e.g., IS393 or IS411) using a beam between \(^{117}\text{Cd}\) and \(^{132}\text{Cd}\) in the central beam line.

6 REFERENCES


2. Nakhone, L. N.; Young, S. D. Environmental Pollution 1993, 82(1), 73-77.


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\(^4\) General Purpose Separator (GPS) Low mass side of the central beam