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# LITTLE FOREST LEGACY SITE - TECHNICAL REPORT

# Radionuclide Sorption Studies of Co, Cs and Sr on LFLS Soils

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# Purpose

The main objectives of this report are to provide:

- a summary of sorption studies of the radionuclides Cs-137, Co-57 and Sr-85 for selected soils from the Little Forest Legacy Site (LFLS).
- a compilation of soil characterisation data, sorption data and estimated distribution coefficients for Cs, Sr and Co for the selected LFLS soils.
- a comparison of the site specific Kd values with historical site data and IAEA data.
- a summary of sorption studies of Co for model mineral components of LFLS soils.

# Related documents

Payne, T.E., **2012**. Background Report on the Little Forest Burial Ground Legacy Waste Site. *ANSTO/E-780*, Australian Nuclear Science and Technology Organisation.

Hankin, S. **2012**. Little Forest Burial Ground - Geology, Geophysics and Well Installation 2009−2010; *ANSTO/E-781*; Vol. 1 and Vol. 2. Australian Nuclear Science and Technology Organisation.

Payne, T.E., **2015**. Little Forest Legacy Site – Summary of Site History until the Commencement of Waste Disposal in 1960. *ANSTO E-782*. Australian Nuclear Science and Technology Organisation.

Payne, T.E., Kinsela, A.S., Bligh, M., Rowling, B., Hughes, C.E., Hankin, S., Anderson, D., Cendon, D., Wilsher, K. and Comarmond, M.J., **2018**. Installation of a Pilot Experimental Trench at the Little Forest Legacy Site. *ANSTO E-786*. Australian Nuclear Science and Technology Organisation.

Johansen, M.P., Payne, T.E., Comarmond, M.J., Harrison, J.J., Blackley, R., and Kabir, A., **2020**. Dose rate estimates to humans and wildlife for a range of potential future scenarios. *ANSTO E-787*. Australian Nuclear Science and Technology Organisation.

# Scope

This document focuses on the sorption studies of Co, Cs and Sr on LFLS materials representing the various lithologies at the site.

# Table of Contents

# 1. Introduction

## 1.1. Background

The Little Forest Legacy Site (LFLS) is located within the ANSTO Buffer Zone Boundary near Sydney in South Eastern Australia. This site, previously known as the Little Forest Burial Ground (LFBG), was used by the former Australian Atomic Energy Commission (AAEC) during the 1960's to dispose of waste containing low levels of radioactivity in a series of shallow trenches which was the accepted international practice at the time [Payne, 2012]. Figure 1.1 shows the location of Little Forest Legacy Site (LFLS) in relation to Australia and the city of Sydney (inset), the ANSTO Buffer Zone boundary, the location of the waste disposal trenches and the local topography. Historically, a number of drilling operations took place at the site since 1958 resulting in monitoring boreholes/wells at the site. There has been regular environmental monitoring of the site since 1966, during which measurable amounts of various radionuclides including Co-60, Sr-90 and Cs-137 have been observed in some soils, groundwater and vegetation samples taken in close proximity to the disposal area [Payne 2012, AAEC 1985].

A comprehensive soil sampling campaign was undertaken in 2009 as part of a research program encompassing field and laboratory studies of the LFLS. Around this time, 33 soil cores (coded CH series) were collected with depths below the ground surface ranging between 1.7 and 5.0 m and drilled using push tube and sectional flight auger (SFA) methods [Hankin, 2012]. The direct push method yielded intact soil cores while the auger technique yielded unconsolidated soil auger chips. These cores were used to provide lithological information and samples for laboratory testwork. A further drilling program was undertaken in 2010 to install additional wells, categorised as the W series [Hankin, 2012].

Geochemical characterisation and sorption studies performed on subsamples from the CH and W series are the subject of this report.

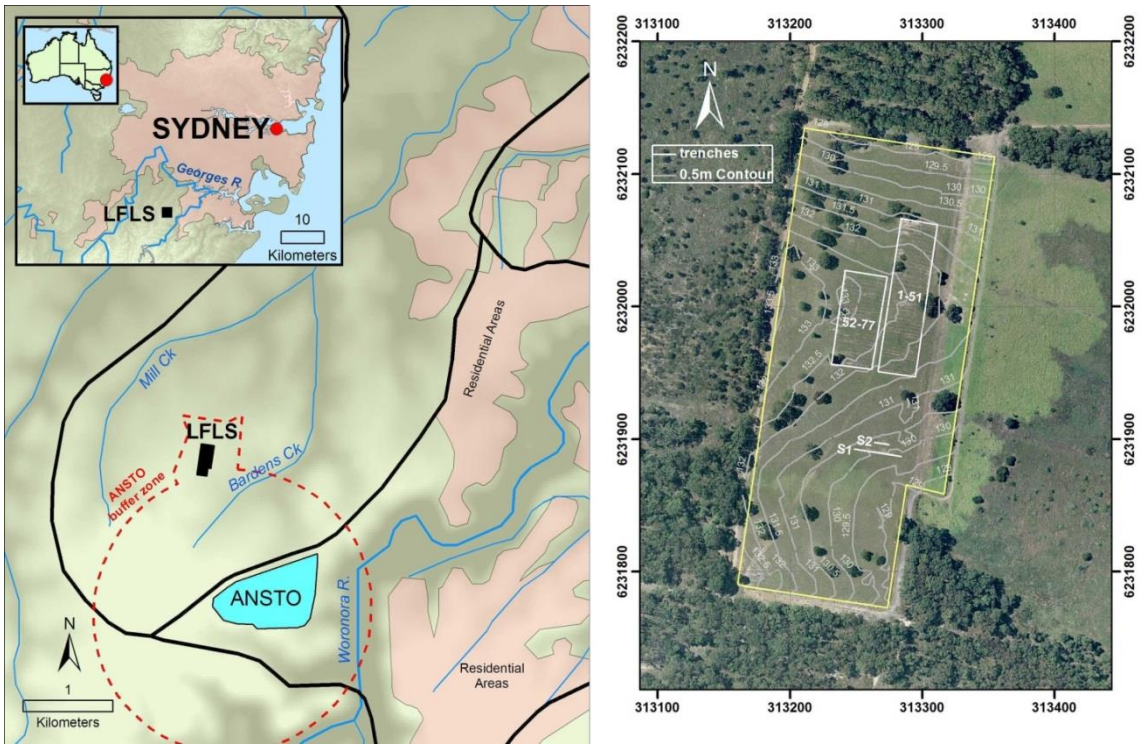


**Figure 1.1: Location of Little Forest Legacy Site (LFLS) in relation to Australia and the city of Sydney and ANSTO (inset). The schematic on the left indicates the ANSTO buffer zone (red dashed line) which includes the LFLS and the photograph on the right is of the LFLS site showing the location of the waste disposal trenches (labelled 1-51, 52-77, S1 and S2) and the local topography [Cendon et al., 2015].**

## 1.2. Rationale for study

The purpose of the present study was to investigate the sorption of the radionuclides Co, Cs and Sr on soil materials sourced from cores sampled at the Little Forest Legacy site. Co, Sr and Cs were chosen as elements of interest because waste containing radioactive isotopes of these elements (primarily Co-60, Sr-90 and Cs-137) are known to have been disposed at this site and are (or have previously been) readily detectable in some environmental samples collected from the site (e.g. groundwater, soils and vegetation). The soil samples were taken at various depth intervals along the core length representing the various lithologies at the site. The sorption studies, coupled with targeted characterization of the samples, will provide insight to the key site characteristics governing contaminant release and transport of these radionuclides at Little Forest. The sorption studies allow for an estimate of site specific distribution coefficient (Kd) values for Co, Sr and Cs under controlled experimental conditions that could be utilised in groundwater fate and transport modelling and dose assessment studies.

# 2. Terminology

| | |
|---|---|
| AAEC | Australian Atomic Energy Commission |
| ANSTO | Australian Nuclear Science and Technology Organisation |
| CEC | Cation Exchange Capacity |
| CH | Corehole |
| CSIRO | Commonwealth Scientific Industry Research Organisation |
| EC | Electrical Conductivity |
| IAEA | International Atomic Energy Agency |
| ICPAES | Inductively Coupled Plasma Atomic Emission Spectrometry |
| ICPMS | Inductively Coupled Plasma Mass Spectrometry |
| Kd | Distribution Coefficient |
| LFBG | Little Forest Burial Ground |
| LFLS | Little Forest Legacy Site |
| OM | Organic Matter |
| SFA | Sectional flight auger |
| XRD | X-ray Diffraction |

# 3. Soil sample selection and sample preparation

## 3.1. Sample selection

### 3.1.1. Location of sites

The samples for sorption studies presented in this report were surface soils and corehole samples obtained from CH1, CH1A, CH30 and W2D site locations indicated in Figure 3.1. These sites represent various locations in the vicinity and well away from the legacy trenches with CH1 and CH1A located at the base and midway between the two sets of legacy trenches and CH30 located further north and near the only trench water sampler in existence at the site of the legacy trenches [Payne, 2013] at this point in time. W2D was located at the south western end of the LFLS, far removed from the legacy trenches. It was noted that materials from this location are likely to be impacted by discharge from the nearby Harrington's Quarry due to groundwater flow paths [Cendon et al., 2015].

### 3.1.2. Sample Description

The samples selected for the testwork are summarised in Table 3.1.

**Table 3.1 Samples for characterisation and sorption studies in the vicinity of the trenches.**

| | CH1 | CH1A | CH30 |
|---|---|---|---|
| **LITHOLOGY** | **Depth (m)** | | |
| TOPSOIL | 0 - 0.2 | | 0 - 0.2 |
| SILTY CLAY | | | 0.2 -0.4 |
| CLAY | | 0.4 -0.6 | 0.4 - 1.0 |
| SHALEY CLAY | | 1.4 - 1.6 | 1.0 - 1.5 |
| SILTSTONE | | | 1.5 - 2.0 |
| SHALE | | 2.4 -2.6 | |
| | | 3.4 -3.6 | |

**Table 3.2 Background samples for characterisation and sorption studies.**

| | W2D |
|---|---|
| **LITHOLOGY** | **Depth (m)** |
| TOPSOIL | 0 - 0.5 |
| SILTY CLAY | 1.0 - 1.5 |
| SHALE | 2.5 - 3.0 |
| CLAY | 4.5 - 5.0 |
| SILTSTONE | 6.5 |
| SHALE | 9.5 |

These samples represent the stratigraphy at the LFLS site, classified into lithological units as described in core logs, namely topsoil, leached/weathered zone shale (further classified as silty clay, clay, shaley clay and siltstone) and parent shale. Figure 3.2 shows a schematic and photograph of CH1A as an example of a typical corehole profile. The core logs for CH1, CH1A, CH30 and W2D are provided in Appendix 1.

## 3.2. Sample preparation

A description of the coring and subsampling of the various cores is provided below. There was some variation in the methods used for subsampling of the various surface soils and cores because of the limited amount of sample available from the drilling programs.

CH1A: The core was acquired by a direct push method. The soil core was split in half lengthways and four 20 cm slices were subsampled from the core as indicated by the arrows in Figure 3.2 to represent the following lithologies:

- Between 0.4 and 0.6 m – clay
- Between 1.4 and 1.6 m – shaley clay/shale
- Between 2.4 and 2.6 m – shale
- Between 3.4 and 3.6 m – shale

CH30: The first two metres of core below the ground surface was obtained using a direct push method. The entire length of this core was subsampled in 10 cm intervals.

W2D: Samples were obtained from grab samples previously collected during the field augering for well construction. Additional sub-samples of bulk material were used for the mineralogical assessment. (see Table 4.2)

CH1 surface soil: The sample was obtained by direct push method.

All samples were air-dried and disaggregated using a rubber mallet and a sub-sample taken for grain size analysis. The remainder was oven dried at 60 ºC prior to sample processing. Coning and quartering and/or a sample riffler were used to provide representative samples with size fractionation (< 2 mm; < 1 mm) facilitated by sieving to be used for sorption and soil characterisation studies. Pulverised sub-samples were prepared using a ring-grinder to be used for elemental assays. All samples were retained in tared plastic sealable bags and stored at ambient temperature.

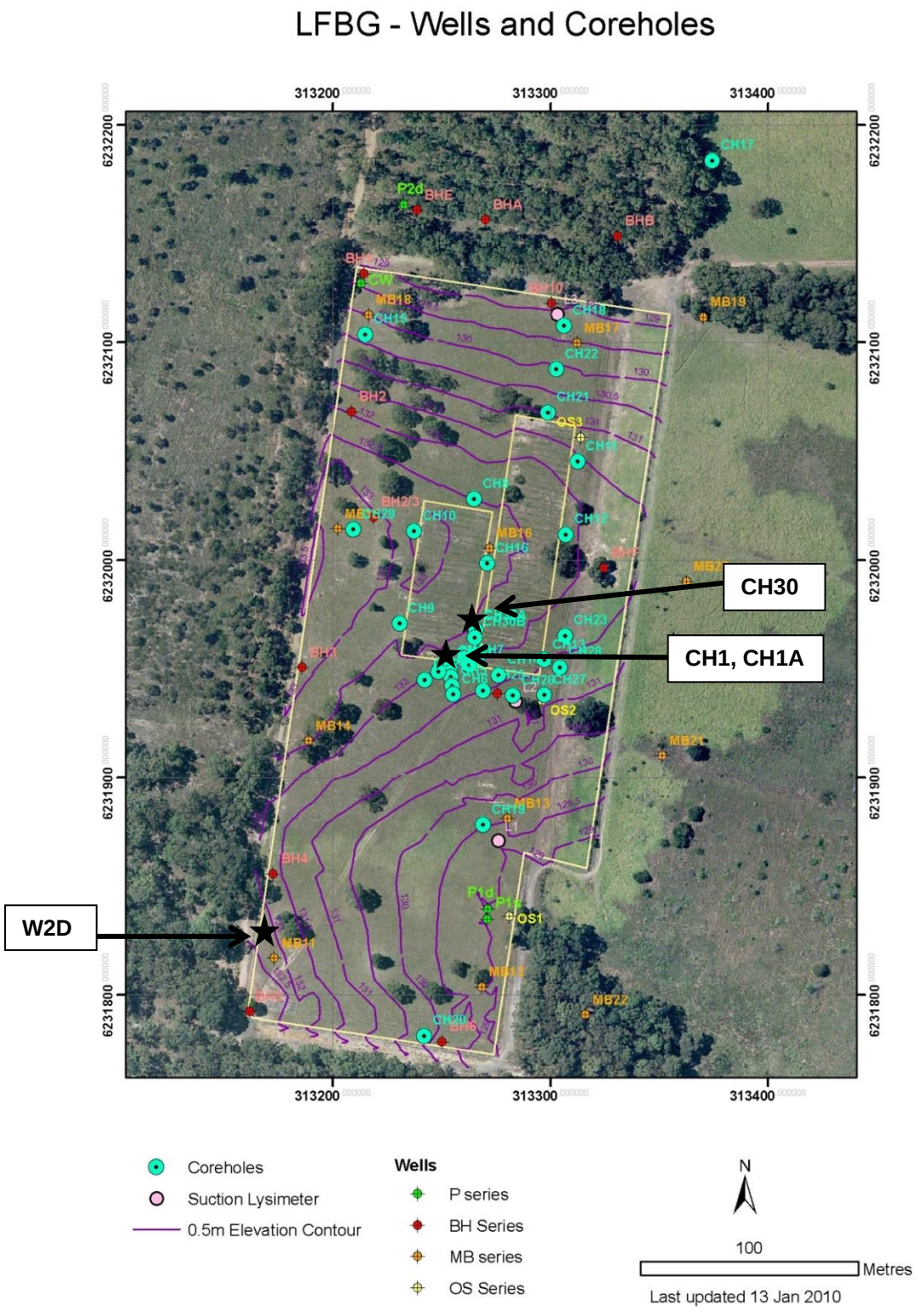


**Figure 3.1: Location of Little Forest Legacy Site (LFLS) samples used for soil characterisation and sorption studies.**

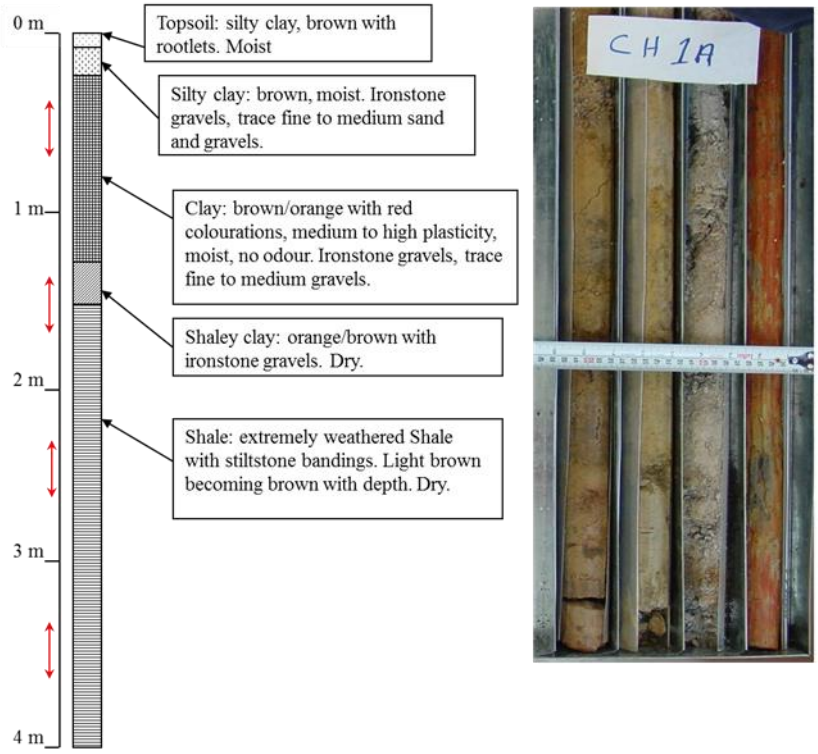


**Fig. 3.2: Schematic of the various lithologies for CH1A and a photograph of the CH1A cores. The core was partitioned in 1 m intervals, with the core at the far right of the photograph representing a sub-section of core obtained from the 0 – 1 m depth, and the core at the far left of the photograph representing a subsection from the 3 – 4 m depth.**

# 4. Geochemical characterisation of soil samples

## 4.1. Characterisation methods

### 4.1.1. pH and EC of 1:5 slurry

The pH and electrical conductivity of soil samples (EC) were conducted on a 1:5 soil/water extract as per the method of Rayment and Higginson [1992]. Non-pulverised samples (5 g) were prepared as slurries and placed in a shaker bath or incubator (150 rpm, at 25 ºC) for one hour. The samples were allowed to settle for 25 minutes prior to measurements of EC and pH, respectively.

### 4.1.2. Cation Exchange capacity

The cation exchange capacities (CEC) of non-pulverised samples were determined using methylene blue and the tetrasodium pyrophosphate method of Wang *et al*. [1996]. Half a gram of soil sample was accurately weighed into a conical flask to which 50 mL of a 20 g/L sodium pyrophosphate solution was added. The slurry was gently boiled on a hot plate for ca. 20 minutes and then allowed to cool at room temperature. The suspension was titrated with a fresh solution of methylene blue (0.05 M or 0.1 M) with 0.5 mL aliquots, using the spot test to determine the end point.

### 4.1.3. Elemental Assay

Elemental assays were conducted on pulverised sub-samples that were subsequently microwave digested in strong acid and analysed by inductively coupled plasma atomic emission spectroscopy (ICP-AES) and inductively coupled plasma mass spectrometry (ICP-MS). The subsamples were digested according to ANSTO method I-2995 - Inorganic: microwave assisted acid digestion and analysed using ANSTO method I-2809 (ICPMS) and ANSTO method I-3775 (ICPAES).

### 4.1.4. Grain size analysis

Two methods were used for grain size analysis. The grain size determinations for the CH1, CH30 and W2D samples were performed using a Mastersizer 2000 [Malvern Instruments Ltd., 2007]. This method is performed for size fractions < 1 mm and provides classification of grain size as sand, silt and clay. Samples were sieved to < 1 mm and the sieved subsamples were initially dispersed in water and ultrasonicated for several minutes prior to analysis on the Mastersizer 2000. The analyses for the CH1A samples were performed by CSIRO [Raven and Self, 2011], with a focus on an assessment of the finer clay fractions. The method used involved repeated dispersion of the bulk sample in 1 M sodium chloride solution followed by centrifugation to recover the coarse clay (> 2 μm) and fine clay (< 2 μm) fractions.

### 4.1.5. BET surface area determination

The specific surface area was determined using the BET-$N_2$ gas adsorption method [IUPAC, 1985] with samples degassed under vacuum at a temperature of 100 ºC prior to the BET surface area determination.

### 4.1.6. Mineralogy

The mineralogical assessment of CH1, CH30 and W2D samples was conducted at ANSTO. Samples were milled in a ring grinder and analysed as pressed powders with random orientation. The powder X-ray diffraction data were obtained with an X'pert Pro diffractometer using Cu $K_{\alpha 1,2}$ radiation with a Ni filter. The PANanalytical analysis package High Score Pro was used for quantitative analysis

The mineralogical assessment of the CH1A samples was performed by CSIRO [Raven and Self, 2011]. Subsamples were micronized for 10 min under ethanol and the resulting slurries oven dried at 60 ºC. These samples were subsequently homogenised in a mortar and pestle and prepared as pressed powders for XRD. The samples showed variable hydration of the interlayer, so were calcium saturated and the data reanalysed.

To recover the < 2 μm and > 2 μm fractions, a subsample of the bulk was prepared by repeated dispersion of the sample in 1 M NaCl as described in Section 4.1.4. The suspensions were flocculated with excess NaCl, treated with acetic acid to remove carbonate minerals, calcium saturated twice (using 1 M $CaCl_2$), washed with water followed by alcohol, with centrifugation at each step. The samples were oven dried at 60 ºC and ground in an agate mortar and pestle and prepared as pressed powders to achieve random orientation of the particles for XRD analysis [Raven and Self, 2011]. The process was also repeated for XRD analysis of the < 0.2 μm and > 0.2 μm sub-samples (randomly oriented).

Oriented specimens of the < 2 μm and < 0.2 μm fraction were prepared by dispersion of the coarse clay and fine clay fractions in deionised water using a high energy ultrasonic probe, and deposition onto 0.2 μm cellulose nitrate filter membranes under suction. The sample was subsequently magnesium saturated using 1 M $MgCl_2$, washed several times with deionised water followed by glycerol and mounted onto a 25 mm aluminium disc using double sided tape.

The XRD patterns were recorded with a PANalytical X'pert Pro Multi-purpose Diffractometer using Fe filtered Co $K_\alpha$ radiation. Quantitative analysis was performed on the XRD data using the commercial package SIROQUANT from Sietronics Ltd and results normalised to 100%.

## 4.2. Soil characterisation results

The geochemical characterisation data obtained for the various subsamples are summarised in Table 4.1.

**Table 4.1 Summary of soil physical and chemical properties**

| COREHOLE | Depth interval (m) | Lithology | size fraction | pH | EC (μS/cm) | CEC (cmol/kg) | BET SA ($m^2/g$) | Elemental Assay | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | Cs (μg/kg) | Co | Sr (mg/kg) |
| CH1 | 0 - 0.2 | topsoil | < 2 mm | 5.48 | 37.0 | nd | 20.0 | nd | nd | nd |
| CH1A | 0.4 -0.6 | clay | < 1 mm | 5.33 | 11.1 | 16.3 | 40.2 | 14 000 | 2 030 | 164 |
| | 1.4 - 1.6 | shaley clay | < 1 mm | 5.14 | 13.7 | 17.3 | 47.1 | 17 000 | 1 090 | 242 |
| | 2.4 - 2.6 | shale | < 1 mm | 5.45 | 8.1 | 7.0 | 36.2 | 14 600 | 1 330 | 206 |
| | 3.4 - 3.6 | shale | < 1 mm | 5.91 | 13.0 | 10.1 | 27.2 | 13 700 | 4 610 | 170 |
| CH30 | 0 - 0.2 | topsoil | < 2 mm | 5.06 | 16.6 | 7.8 | 18.3 | 10460 | 2300 | 136 |
| | 0.2 - 0.4 | silty clay | < 2 mm | 5.53 | 22.1 | 9.3 | 15.1 | 9360 | 2740 | 81.5 |
| | 0.4 - 0.6 | clay | < 2 mm | 5.50 | 21.7 | 14.6 | 20.6 | 12400 | 226 | 145 |
| | 0.6 - 0.8 | clay | < 2 mm | 5.03 | 18.1 | 16.3 | 45.2 | 13600 | 1420 | 191 |
| | 0.8 - 1.0 | clay | < 2 mm | 5.27 | 13.3 | 15.2 | 38 | 11000 | 1310 | 142 |
| | 1.0 - 1.2 | shaley clay | < 2 mm | 5.25 | 12.1 | 5.6 | 24.1 | 11300 | 3900 | 200 |
| | 1.2 -1.4 | shaley clay | < 2 mm | 5.07 | 11.0 | 4.0 | 23 | 10450 | 1685 | 179 |
| | 1.4 - 1.6 | shaley clay/siltstone | < 2 mm | 5.74 | 8.5 | 3.6 | 25.8 | 12000 | 895 | 192 |
| | 1.6 - 1.8 | siltstone | < 2 mm | 5.56 | 8.1 | 7.8 | 25.5 | 11500 | 11400 | 184 |
| | 1.8 - 2.0 | siltstone | < 2 mm | 5.29 | 10.7 | 10.0 | 25.8 | 11350 | 11125 | 192 |
| W2D | 0 - 0.5 | topsoil | < 2 mm | 5.44 | 24.1 | nd | 71.9 | nd | nd | nd |
| | 1.0 - 1.5 | silty clay | < 2 mm | 4.84 | 25.9 | 11.1 | 45.5 | 7860 | 1950 | 98.4 |
| | 2.5 - 3.0 | shale | < 2 mm | 5.21 | 23.0 | 7.2 | 32.4 | 6750 | 2800 | 167 |
| | 4.5 - 5.0 | clay | < 2 mm | 5.27 | 34.8 | 6.9 | 14.6 | 5290 | 5420 | 214 |
| | 6.5 | siltstone | < 2 mm | 6.83 | 41.2 | 3.9 | 9.1 | 1670 | 15700 | 125 |
| | 9.5 | shale | < 2 mm | 6.88 | 50.9 | 5.5 | 11.3 | 32700 | 23600 | 288 |

nd = not determined

### 4.2.1. Soil pH and EC

The pH of the soils for core depths up to 5 m typically ranges from 4.9 to 5.9. The deeper samples of W2D (depth greater than 5 m) are near neutral (soil pH 6.6 to 6.8) and this is attributed to the elevated siderite content in those samples as determined from mineralogical assessment (see Section 4.2.5 below).

The W2D samples have higher electrical conductivities (EC) ranging from 23 to 51 μS/cm in comparison with the CH1A and CH30 samples. The EC for subsamples of the CH1A and CH30 cores are similar along the core profile, ranging from 8 to 22 μS/cm. The EC for W2D samples typically increase with depth and the elevated values may arise from the impact of leachate flow from the nearby Harrington's Quarry [Cendon et al., 2015] as well as from the dissolution of carbonates inherent in these materials.

### 4.2.2. Cation exchange capacity

The cation exchange capacities (CEC) for CH1A, CH30 and W2D soils are reported in Table 4.1 and range from 7.0 to 17.3 cmol $kg^{-1}$ for the CH1A samples, 3.6 to 16.3 cmol $kg^{-1}$ for the CH30 samples, and 3.9 to 11.1 cmol $kg^{-1}$ for the W2D samples. CECs determined for soil samples from LFLS have previously been reported with values ranging from 7.1 to 9.0 cmol $kg^{-1}$ as shown in Table 4.2 [ Isaac and Mears, 1977]. The four soils from the earlier study contained more than 50% kaolinite and have low CEC values. This is consistent with kaolinitic soils where kaolinite as the dominant clay has a low ability to exchange ions [Goldberg et al., 2005].

**Table 4.2: Description and cation exchange capacity of a soil profile at LFLS (reproduced from Isaac and Mears, 1977).**

| Fraction | Sampling depth (m) | Description | Composition | CEC (cmol $kg^{-1}$) |
|---|---|---|---|---|
| F1 | 0.0-0.5 | topsoil | Kaolinite (>50%)<br>Mica, smectite Quartz (<50%) | 7.3 |
| F2 | 0.5-1.0 | Red-brown clay | Kaolinite (80%)<br>Mixed-layer mica –smectite, Quartz (20%) | 9.0 |
| F3 | 1.0-1.5 | Greyish clay | Kaolinite (80%)<br>Mixed-layer mica-smectite, Quartz (20%) | 7.1 |
| F4 | 1.5-2.5 | Weathered shale | Kaolinite (65%)<br>Micaceous clay mineral (35%) | 8.3 |

### 4.2.3. Elemental Assays for Cs, Co and Sr inherent in samples

Results of the chemical assays for Cs, Co and Sr inherent in the various subsamples are summarized in Table 4.1. With the exception of the W2D shale material sourced at ca 9.5 m below the ground surface, the samples of CH1A and CH30 are more elevated in inherent Cs content than the W2D samples. The Cs content in these samples range from 9.3 ppm to 17 ppm and are significantly higher than the average crustal abundance value of 2 ppm and the global average shale value for Cs of 7 ppm [Krauskopf, 1983; Rudnick and Gao, 2014] whereas the inherent Cs for the W2D samples ranges from 1.6 to 7.9 ppm.

The inherent Co in the samples ranges from 230 to 23 600 ppb, with the majority of samples well below both the average crustal abundance of 26 600 ppb and the global average shale value of 20 000 ppb. The siltstone samples from CH30 and the siltstone and shale samples from W2D have significantly higher Co content (> 10 000 ppb) with the W2D shale sample below 9.5 m exceeding the global average shale value.

The inherent Sr in the samples ranges from 82 to 290 ppm. These values fall well below the average crustal abundance for Sr (320 ppm) and the global average shale value for Sr (400 ppm).

### 4.2.4. Grain Size and BET surface areas

The grain size distribution for subsamples of CH30 is shown in Figure 4.1. Sieving established that up to ca 30% of the particles are > 1 mm when fractionating samples with a maximum particle size of 2 mm. The clay and silt fraction were found to be highest for the first meter below the ground surface, followed by a decrease in clay fraction below 1 m. For grain size analysis of the < 1 mm fraction using the Mastersizer, fluctuating results were obtained on replicate sub-samples when using different ultrasonication times, suggestive that water may not have been the best dispersant for clays and/or that ultrasonication was not suitable for disaggregating clay fractions. These aspects were not explored further in the present study.

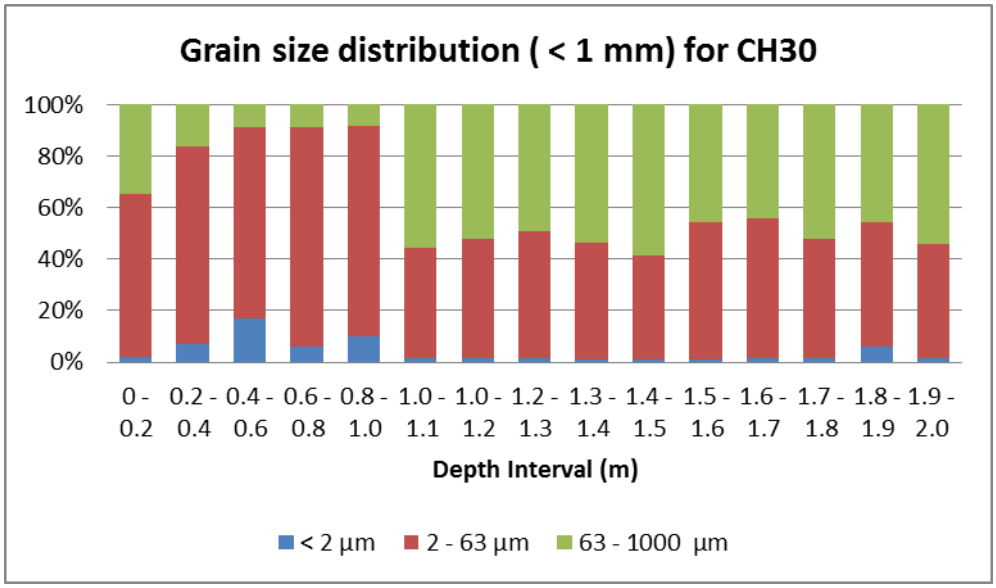


**Fig. 4.1: Grain size distribution for CH30 (< 2 mm).**

The grain size distribution for CH1A was performed by CSIRO using a different method, with greater emphasis on the distribution of coarse (< 2 μm) and fine clay (< 0.2 μm) fractions. Figure 4.2 shows that both the coarse and fine clay fractions, respectively, decrease with depth for the core profile. The clay lithology sample (0.4 – 0.6 m) had significantly more coarse and fine fraction clay (< 2 μm) compared to the shaley clay and shale samples. There is also a decrease in fine fraction clay with depth.

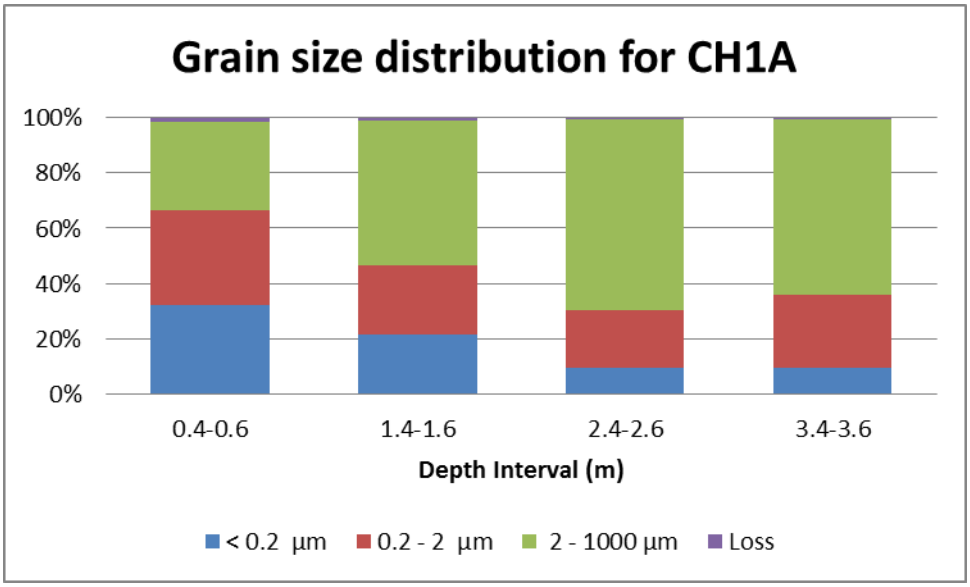


**Fig 4.2: Distribution of grain size in bulk samples (< 1 mm). The coarse clay fraction is represented by the 0.2 – 2 µm fraction, while the fine clay fraction is the fraction below 0.2 µm.**

The BET surface areas for samples of CH1A range between 27 and 47 $m^2/g$ while the BET surface areas for the CH1 topsoil in the near vicinity was somewhat lower at 20 $m^2/g$. The BET surface areas for the CH30 samples range from 15 to 45 $m^2/g$. The BET surface area for samples of W2D range from 9.1 to 72 $m^2/g$ with the highest BET observed for the topsoil sample.

### 4.2.5. Mineralogy

A summary of the mineralogical assessment of the samples is given in Table 4.3 with the distribution of minerals in each sample shown in Figures 4.3 – 4.12.

The CH1 sample is a topsoil/fill with ca. 63% quartz and 34% clay and mica minerals (predominantly kaolinite and mixed layer clays) with trace amounts of anatase, rutile and goethite (each < 1%) (Figures 4.3 and 4.4).

The CH1A samples comprise predominantly of quartz (25-31%), interstratified illite-smectite (23-35%) and kaolinite (28-31%) of similar proportions, with *ca*. 6-11% mica and/or illite. The interstratified illite-smectite has been estimated to be composed of approximately 70% illite and 30% smectite layers. Smectite (4-8%) was also found in the uppermost clay layers [Raven and Self, 2011]. Trace anatase and iron oxides were found in the majority of samples. XRD analysis of the < 1 mm, < 2 µm and < 0.2 µm fractions of the CH1A soils (Figs. 4.4 to 4.6) show that kaolin and interstratified illite-smectite and smectite occur predominantly in the finest clay fractions.

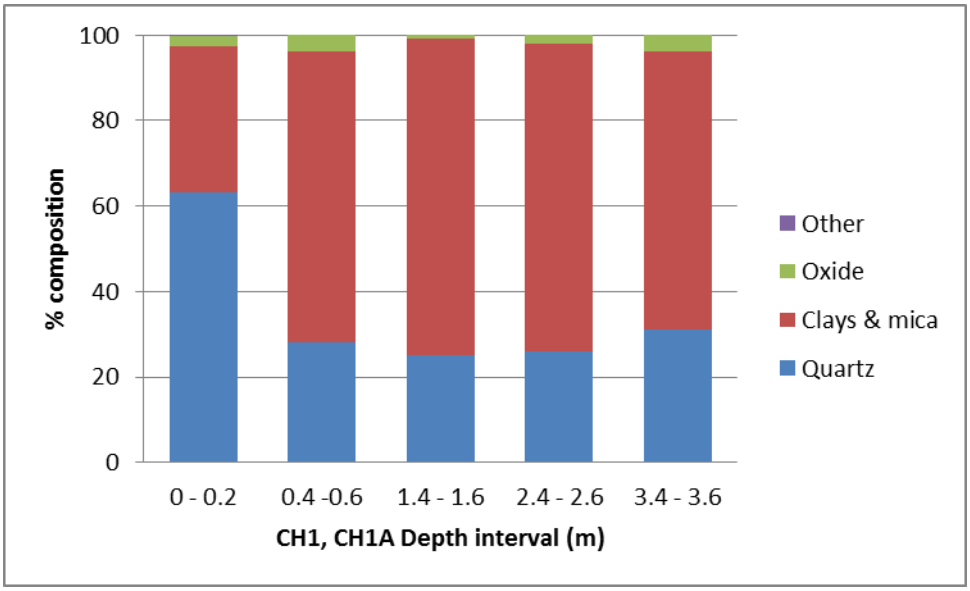


**Figure 4.3: Percentage composition of quartz, clays & mica and oxides in corehole samples of CH1 and CH1A for various depth intervals.**

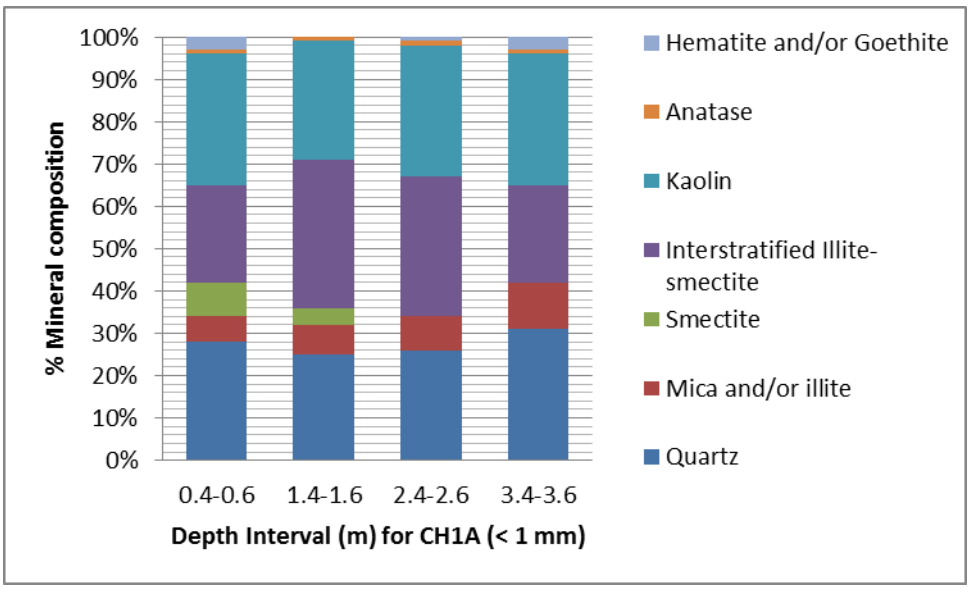


**Figure 4.4: Mineralogical composition of the < 1 mm fraction of CH1A corehole samples at various depth intervals as determined by XRD analysis.**

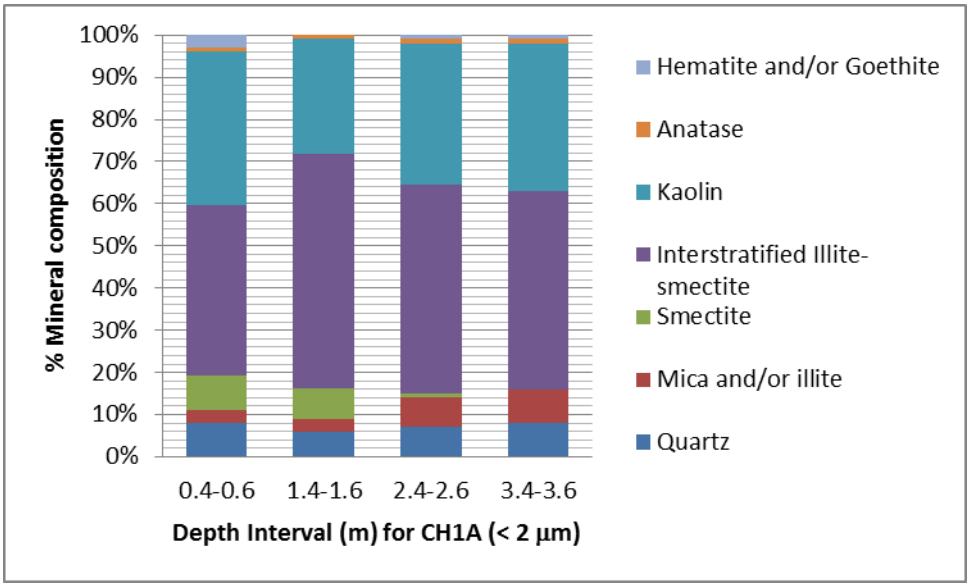


**Figure 4.5: Mineralogical composition of the < 2 µm fraction of CH1A corehole samples at various depth intervals as determined by XRD analysis.**

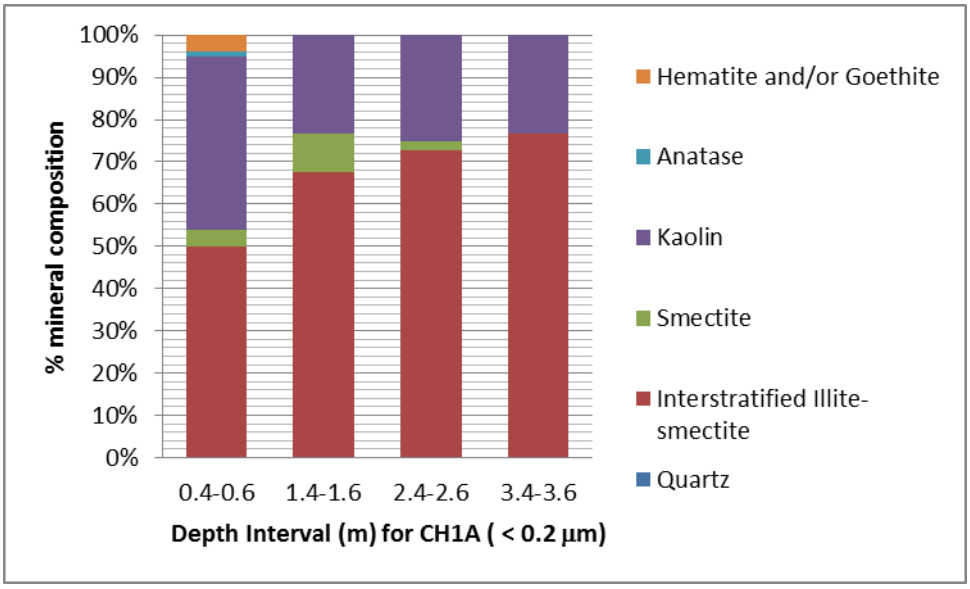


**Figure 4.6: Mineralogical composition of the < 0.2 µm fraction of CH1A corehole samples at various depth intervals as determined by XRD analysis.**

Figure 4.7 and 4.8 show the distribution for quartz, clays and mica, and oxides for the < 2 mm and < 1 mm samples of CH30, respectively. The mineralogical distribution is similar for sub-samples at the two particle size fractions for these mineral groups. The CH30 samples are higher in quartz content (40 – 60%) relative to the CH1A samples, with clays and mica making up the bulk of the difference. These samples also have oxide content of up to *ca*. 10%, with both anatase and goethite pervasive throughout the 2 m depth profile. The mineralogical composition for the CH30 samples is shown in Figures 4.9 and 4.10.

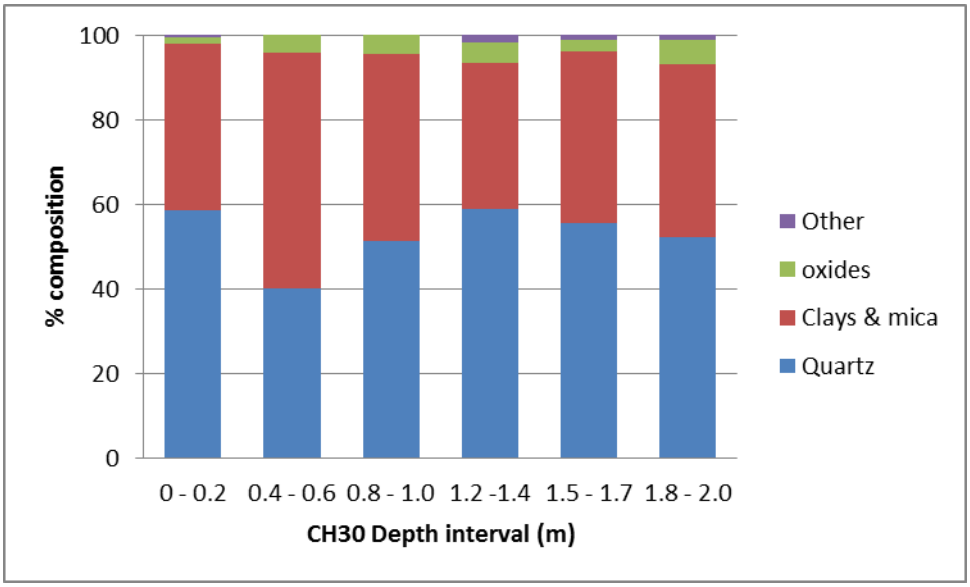


**Figure 4.7: Percentage composition of quartz, clays & mica and oxides in corehole samples of CH30 (< 2 mm) for various depth intervals.**

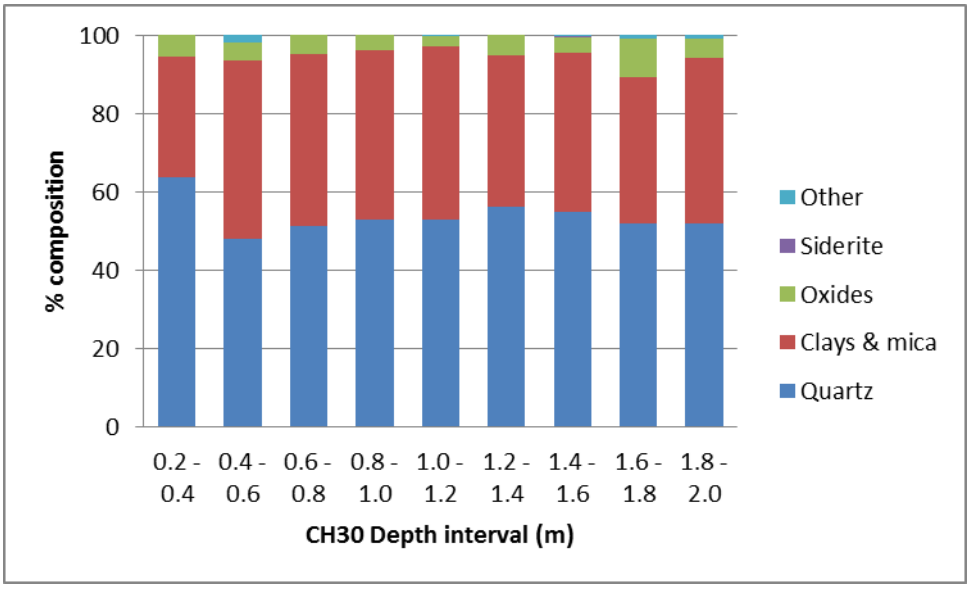


**Figure 4.8: Percentage composition of quartz, clays & mica and oxides in corehole samples of CH30 (< 1 mm) for various depth intervals.**

**Table 4.3: Mineralogical assessment of LFLS soil samples from coreholes CH1, CH1A, CH30 and W2D.**

| COREHOLE | Lithology | Depth interval (m) | Mid-point (m) | size fraction | TECTOSILICATES | | PHYLLOSILICATES | | | | | | | | OXIDES/HYDROXIDES | | | | CARBONATES |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | Quartz & Feldspars (%) | | Clay & Mica minerals (%) | | | | | | | | Ti oxides (%) | | Fe oxide/hydroxides (%) | | (%) |
| | | | | | Qtz | Orthoclase | Kaolin | Dickite | Mixed Layer | Illite | Smectite | Mica and/or illite | Muscovite | Biotite | Rutile | Anatase | Goethite | Hematite | Siderite |
| CH1 | topsoil | 0 - 0.2 | 0.1 | < 2 mm | 63.1 | | 17.3 | 1.7 | 15 | | | | | 0.2 | 0.8 | 0.7 | 0.9 | | |
| CH1A | clay | 0.4 -0.6 | 0.5 | < 1 mm | 28 | | 31 | | 23 | | 8 | 6 | | | | 1 | 3 | | |
| | shaley clay | 1.4 - 1.6 | 1.5 | < 1 mm | 25 | | 28 | | 35 | | 4 | 7 | | | | 1 | | | |
| | shale | 2.4 - 2.6 | 2.5 | < 1 mm | 26 | | 31 | | 33 | | | 8 | | | | 1 | 1 | | |
| | shale | 3.4 - 3.6 | 3.5 | < 1 mm | 31 | | 31 | | 23 | | | 11 | | | | 1 | 3 | | |
| CH30 | topsoil | 0 - 0.2 | 0.1 | < 2 mm | 58.6 | 0.4 | 23.7 | 8.1 | 7.3 | | | | | 0.2 | 0.3 | 1.3 | 0 | 0 | |
| | | | | < 1 mm | 58.4 | 0.2 | 17.2 | 9.1 | 9.5 | | | | | 1.2 | 0.9 | 1.4 | 2 | 0.3 | |
| | silty clay | 0.2 - 0.4 | 0.3 | < 1 mm | 63.8 | 0.2 | 6.1 | 5.8 | 18.4 | | | | | 0.4 | 1.6 | 1.3 | 1.1 | 1.5 | |
| | clay | 0.4 - 0.6 | 0.5 | < 2 mm | 40 | 0 | 12.2 | 6.1 | 37.5 | | | | | 0 | 0 | 0.8 | 3.3 | 0 | |
| | clay | | | < 1 mm | 47.9 | 0.2 | 11.9 | 4.2 | 29.4 | | | | | 0 | 0.2 | 1.3 | 3.3 | | |
| | clay | 0.6 - 0.8 | 0.7 | < 1 mm | 51.1 | 0 | 10.9 | 10.7 | 22 | | | | | 0.3 | 0 | 1.3 | 3 | 0.6 | |
| | clay | 0.8 - 1.0 | 0.9 | < 2 mm | 51.4 | 0 | 27.6 | 7 | 9.4 | | | | | 0.1 | 0 | 1.5 | 2.4 | 0.5 | |
| | clay | | | < 1 mm | 52.8 | 0 | 20.8 | 6.9 | 15.4 | | | | | 0.2 | 0.1 | 1.6 | 2 | 0.2 | |
| | shaley clay | 1.0 - 1.2 | 1.1 | < 1 mm | 52.8 | 0.1 | 15.7 | 4.4 | 23.8 | | | | | 0.3 | 0 | 1.1 | 1.4 | 0.2 | |
| | shaley clay | 1.2 -1.4 | 1.3 | < 2 mm | 58.9 | 2 | 19.6 | 6.6 | 7.6 | | | | | 0.7 | 0.4 | 1.6 | 2.7 | 0 | |
| | shaley clay | | | < 1 mm | 56.2 | 0 | 17.6 | 5.2 | 15.4 | | | | | 0.4 | 0.3 | 1.3 | 2.7 | 0.9 | |
| | shaley clay/siltstone | 1.4 - 1.6 | 1.5 | < 1 mm | 54.9 | 0.2 | 16.7 | 6.8 | 16.5 | | | | | 0.5 | 0.2 | 1 | 2.1 | 0.8 | 0.3 |
| | siltstone | 1.5 - 1.7 | 1.6 | < 2 mm | 55.7 | 1.3 | 20 | 5 | 14.8 | | | | | 0.6 | 0 | 1.2 | 1.4 | 0 | |
| | siltstone | 1.6 -1.8 | 1.7 | < 1 mm | 51.9 | 1.9 | 8.1 | 10.3 | 17.9 | | | | | 1.1 | 0.7 | 1.1 | 6 | 1.8 | |
| | siltstone | 1.8 - 2.0 | 1.9 | < 2 mm | 52.3 | 1.1 | 19.5 | 5.5 | 15.2 | | | | | 0.5 | 0.4 | 1.5 | 3.3 | 0.6 | |
| | siltstone | | | < 1 mm | 52 | 0.3 | 18.2 | 5.2 | 18.4 | | | | | 0.3 | 0 | 1.2 | 3.6 | 0 | |
| W2D | topsoil | 0 - 0.5 | 0.25 | < 2 mm | 29.1 | | 33.4 | 1.8 | 1.3 | | | | | | | 1.8 | 32.6 | | |
| | sitly clay | 0.5 -1.0 | 0.75 | bulk | 28.9 | | 38.6 | 11.4 | | 4 | | | 2.4 | | 1 | 2.1 | 10.6 | | 1.1 |
| | shaley clay | 2.0 - 2.5 | 2.25 | bulk | 55.1 | | 25.6 | 2.5 | | 4.6 | | | 8.5 | | 0.7 | 0.8 | 1.7 | | 0.6 |
| | shaley clay | 4.0 - 4.5 | 4.25 | bulk | 57.9 | | 24.3 | 3 | | 3.3 | | | 6.2 | | 0.5 | 0.4 | 4.3 | | 0.1 |
| | clay | 4.5 - 5.0 | 4.75 | bulk | 61 | | 21.7 | 1.8 | | 4.5 | | | 7.6 | | 0.4 | 0.7 | 2.3 | | 0.2 |
| | siltstone | 6 | 6 | bulk | 68.1 | | 11.9 | 4.9 | | 2.1 | | | 4.7 | | 0.5 | 0.2 | 1 | | 6.6 |
| | siltstone | 7.5 | 7.5 | bulk | 72.2 | | 11.4 | 4.4 | | 2.3 | | | 3.9 | | 0.3 | 0.2 | 0.4 | | 4.8 |
| | shale | 10 | 10 | bulk | 51.7 | | 15.2 | 2.9 | | 5.7 | | | 6.9 | | 0.3 | 1.1 | 0.5 | | 15.8 |
| | shale | 10.5 | 10.5 | bulk | 58.1 | | 15.4 | 3.3 | | 4.1 | | | 6.3 | | 0.2 | 0.8 | 1.1 | | 10.9 |

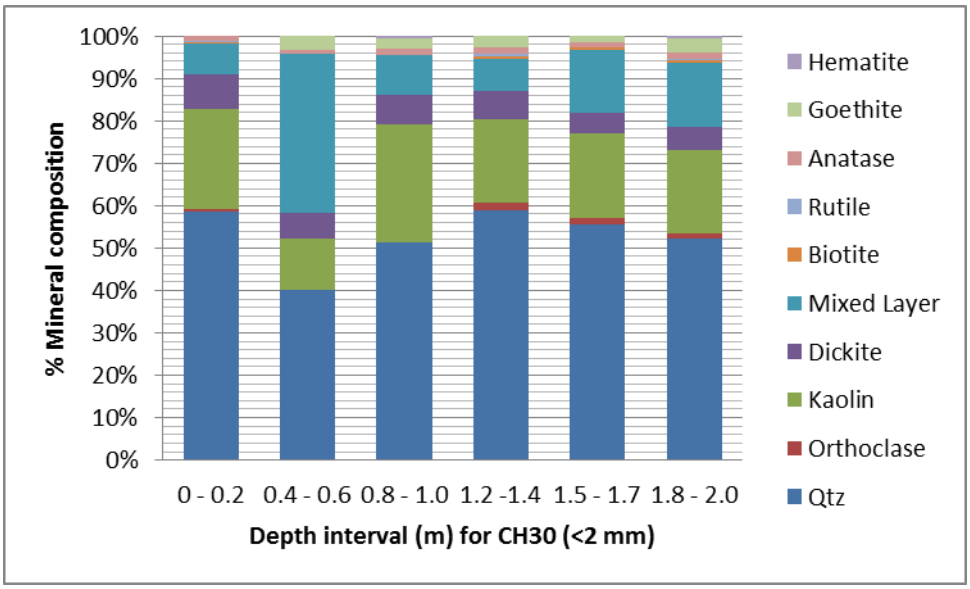


**Figure 4.9: Mineralogical composition of the < 2 mm fraction of CH30 corehole samples at various depth intervals determined by XRD analysis.**

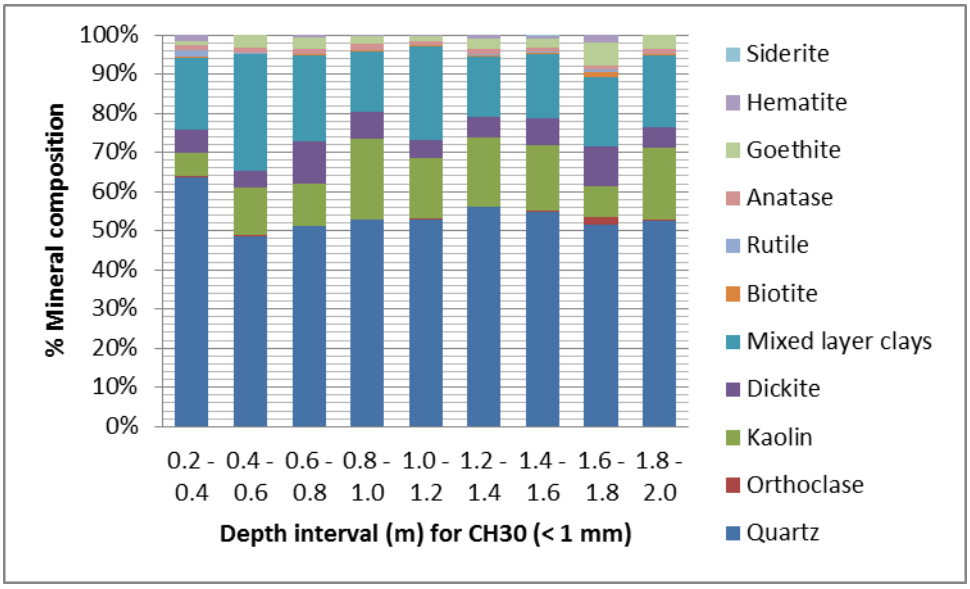


**Figure 4.10: Mineralogical composition of the < 1 mm fraction of CH30 corehole samples at various depth intervals determined by XRD analysis.**

With the exception of the W2D topsoil, the mineralogical assessment of the W2D samples were performed on subsamples of the bulk material (i.e. subsamples were not sieved to recover only the < 2 mm fraction). The W2D samples have ca. 30% quartz in the first meter of core below the ground surface. Below two meters from the ground surface, the quartz content ranges from 50 – 70% with depth. The oxide content is significant in the top meter ranging from 18 – 35%. Siderite is also prevalent at depths below 6 m, ranging from 5 to 18%. The presence of siderite is supported by the higher natural pH observed for soils at the 6.5 and 9.5 m depth interval (Section 4.2.1).

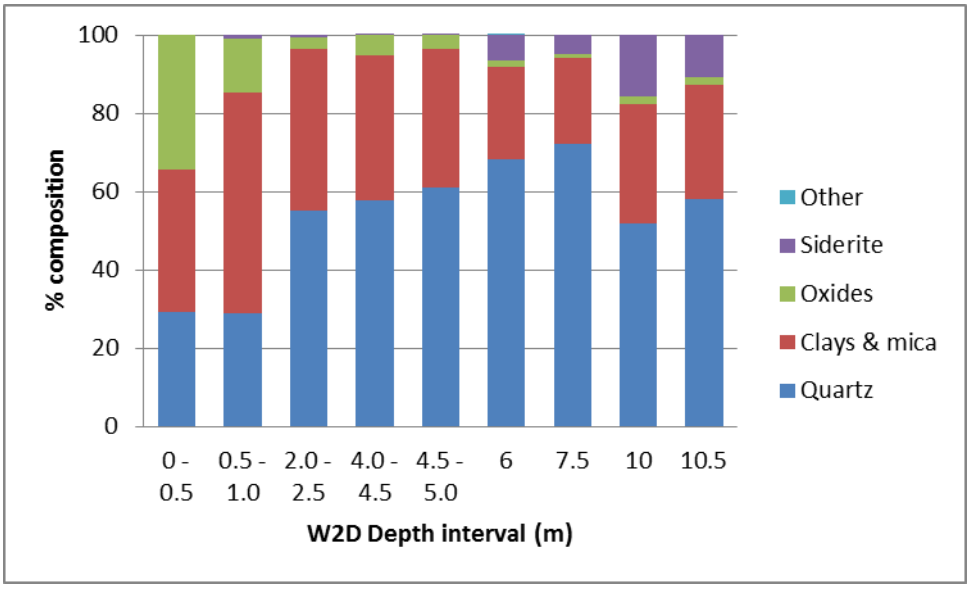


**Figure 4.11: Percentage composition of quartz, clays & mica and oxides in W2D samples for various depth intervals.**

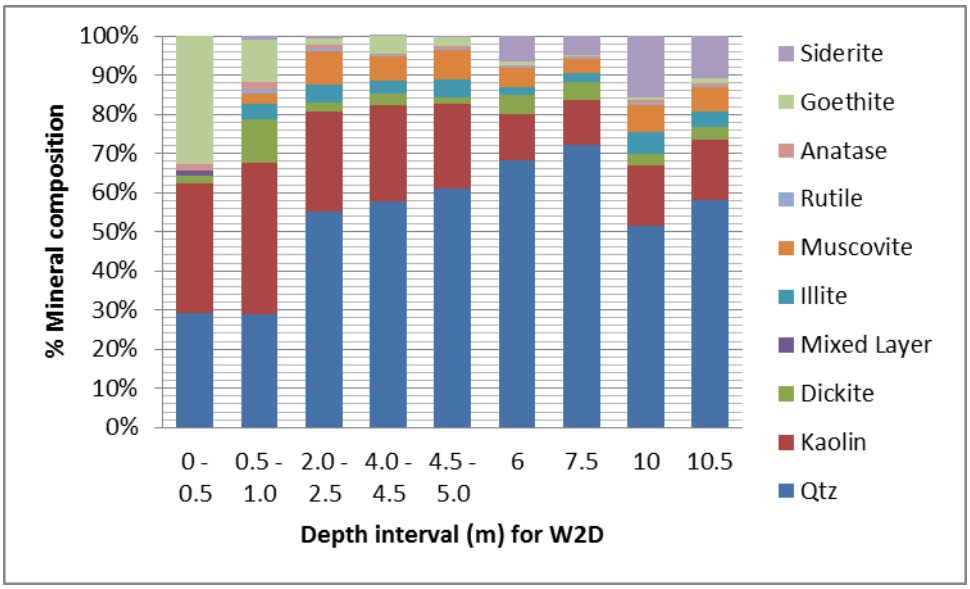


**Figure 4.12: Mineralogical composition of the W2D samples at various depth intervals as determined by XRD analysis.**

The present data shows that the mineralogy across the site is heterogeneous. While it is noted that CH1A has lower amounts of quartz compared to the other locations, the mineralogical data for CH1A presents a better estimate of the phase identification of the clays as orientation of the samples and pretreatment were used to purposefully identify the clays. The higher values for quartz for the other LFLS samples are more typical of estimates in the Sydney Basin for shale lenses within the Hawkesbury sandstone (per comm – Cendon, June 2016).

Early records describe the soils of the site as highly weathered and derived from the shale layer which is generally weathered to a depth of 6 m, resulting in a profile from surficial red-brown clay soil, through mottled grey and silty kaolinite clay with numerous fine sandy partings. Immediately below the soil and weathered material lies light grey leached silty shale grading to fresh dark carbonaceous parent shale with silty interbedding [Isaac and Mears, 1977]. Early records also report the clay in the Lucas Heights area is likely to average 40% $SiO_2$, 30% kaolinite, and 30% illite [Bradd, 2003]. An assessment of the content of these major mineral groups for the clays in the present study is summarized in Table 4.4. The data in Table 4.4 clearly shows the heterogeneity between clay samples both vertically and laterally at the LFLS site.

**Table 4.4: Major mineral groups in LFLS clays from CH1A, CH30 and W2D cores**

| COREHOLE | Depth (m) | Number of samples | $SiO_2$ (%) | Kaolin/dickite (%) | Illite/ smectite/mica /muscovite (%) |
|---|---|---|---|---|---|
| CH1A | 0.4 - 1.6 | 2 | 25 - 28 | 28-31 | 37 - 46 |
| CH30 | 0.2 - 1.6 | 10 | 40 - 64 | 12 - 34.6 | 7.6 - 37.5 |
| W2D | 0.5 - 5 | 4 | 29 - 61 | 23.5 - 50 | 6.4 - 13 |

# 5. Sorption studies of Co, Cs and Sr onto soils

## 5.1. Radiotracers

The radiotracers Cs-137, Co-57 and Sr-85 were purchased from Eckert & Ziegler as carrier-free solutions. Isotopic stock solutions were diluted with 0.1 M HCl to prepare 10 kBq $mL^{-1}$ working solutions.

## 5.2. Batch sorption method

The batch sorption studies were performed in Nalgene centrifuge tubes with a 2 mm hole in the lid to allow air equilibration. These experiments were conducted on selected materials with a mass loading of 10 g/L, an initial activity of *ca.* 500 Bq of each radiotracer and ionic strength of 0.01 M (NaCl). All experiments were conducted at 25 ºC. To investigate the effect of ionic strength on sorption, sorption studies were also conducted on four samples from CH1A at an ionic strength of 0.1 M NaCl. NaCl was chosen as the matrix as most of the groundwater in the vicinity of the trenches at LFLS is of the Na-Cl type [Cendon et al., 2015].

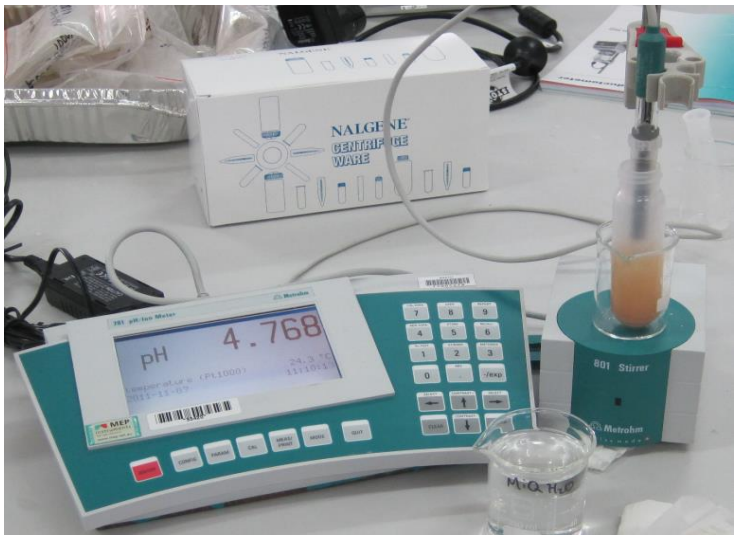


**Fig. 5.1: Set-up for pH determination of a sorption sample**

The solid and aqueous phases were equilibrated for a minimum of 16 h in a temperature controlled shaking water bath at the desired equilibrium pH (target pH) prior to the addition of tracer. In some cases, the equilibration time was extended for much longer periods (several days) to ensure a stable target pH prior to addition of radionuclide tracer. For some samples, the solid and aqueous phases were initially pre-equilibrated without any pH adjustment for several days prior to adjustment to the target pH and subsequent equilibration. After the pre-equilibration at the target pH, the radionuclide tracer was subsequently added and the pH adjusted back to the target pH. The experimental equilibration time was 48 h however, the pH was checked after 24 h and pH adjusted to the target pH, if required, by addition of dilute NaOH or HCl. A combined pH electrode (Metrohm) was used for pH measurements (Fig. 5.1). At completion of the radionuclide equilibration, the final pH was determined for each sorption sample. The aqueous phase was separated by high speed centrifugation at 8000 g for 20 minutes and a 10 mL aliquot subsampled and subsequently acidified to pH < 2 prior to analysis.

## 5.3. Analysis of sorption samples

### 5.3.1. Gamma analysis

The Co, Sr and Cs activities were measured by gamma spectrometry using an ORTEC High-Purity Germanium (HPGe) n-type reduced background detector coupled to an ORTEC DSPEC Pro with MAESTRO software.

### 5.3.2. ICP analysis

Following gamma counting, the aqueous sub- samples were analysed for major metals and minor elements of interest by Inductively Coupled Plasma - Atomic Emission Spectrometry (ICP-AES) and Inductively Coupled Plasma - Mass Spectrometry (ICP-MS).

## 5.4. Sorption results for Co, Cs and Sr

### 5.4.1. Sorption of Co, Cs and Sr for all LFLS samples

The sorption curves for Co, Cs and Sr as a function of pH and at an ionic strength of 0.01 M (NaCl) are shown in Figure 5.2 for over 70 samples of LFLS soils. A compilation of sorption data can be found in Appendix 2. The sorption of Cs typically ranges from 80 to 100 % for LFLS materials and is pH independent whereas the sorption of Co and Sr are pH dependent with a sorption edge between pH 4 and 6. At pH > 6, the sorption of Co nears maximum (i.e. 100%) for the majority of samples, while the sorption of Sr typically ranges from 50 to 90%.

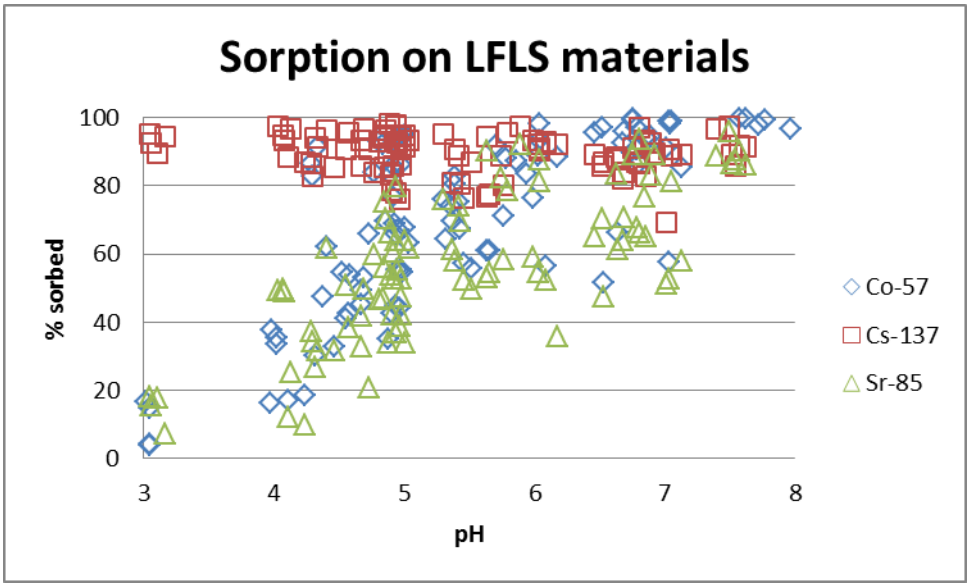


**Figure 5.2: Sorption curves for sorption of Co, Cs and Sr onto topsoil and unconsolidated LFLS materials (soils) as a function of pH. Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

### 5.4.2. Sorption curves as a function of lithologies

Figure 5.3 to 5.9 show the sorption curves obtained for Co, Sr and Cs for the various lithological units of LFLS soils. The pH dependence for Co and Sr is most apparent for the LFLS clays (i.e. silty clays, clays and shaley clays) and the shale material.

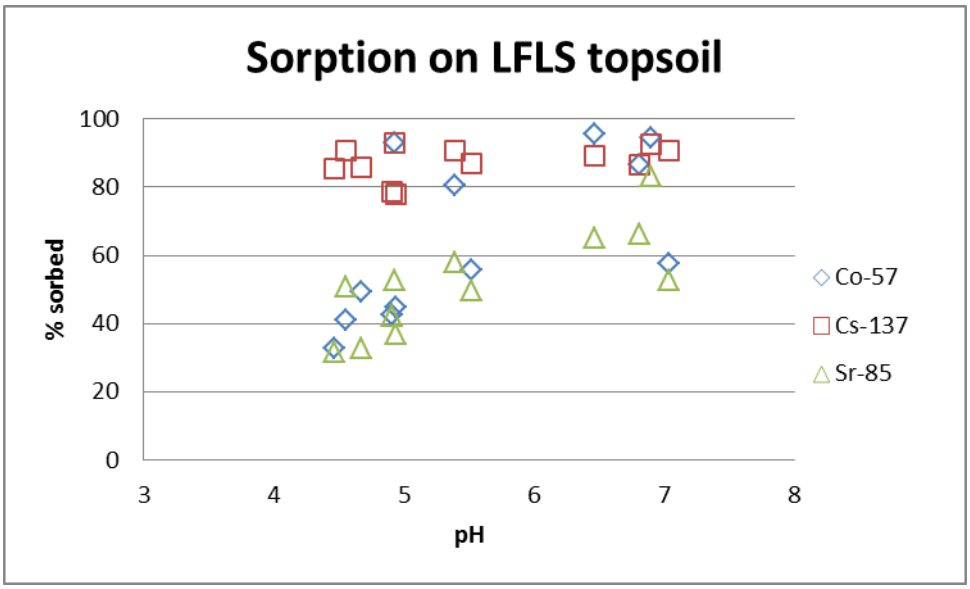


**Figure 5.3: Sorption curves for sorption of Co, Cs and Sr onto LFLS topsoils CH1 (0 – 0.20 m), CH30 (0 – 0.20 m) and W2D (0 – 0.5 m) as a function of pH. Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

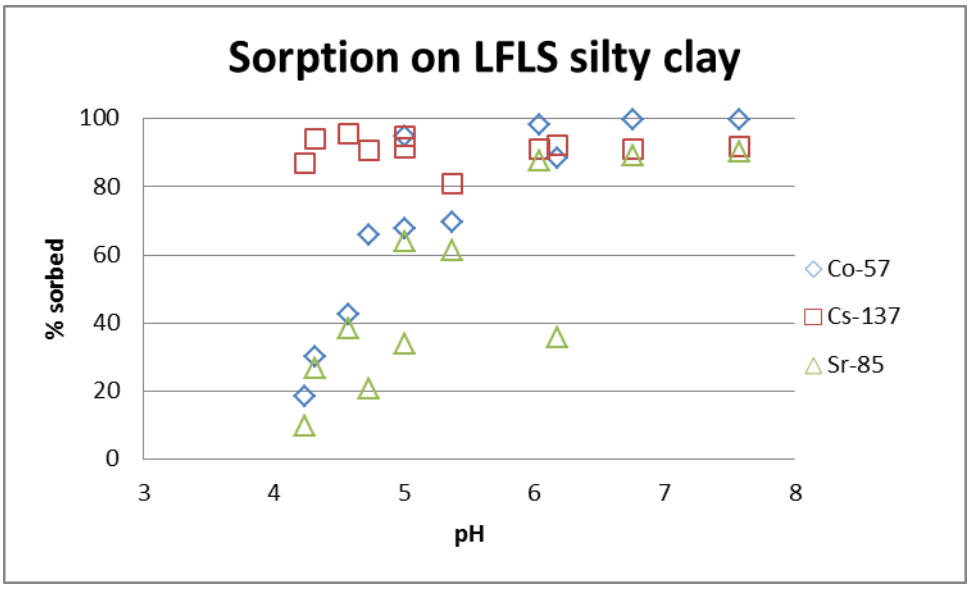


**Figure 5.4: Sorption curves for sorption of Co, Cs and Sr onto LFLS silty clays CH30 (0.2 – 0.4 m); W2D (1.0 – 1.5 m) as a function of pH. Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

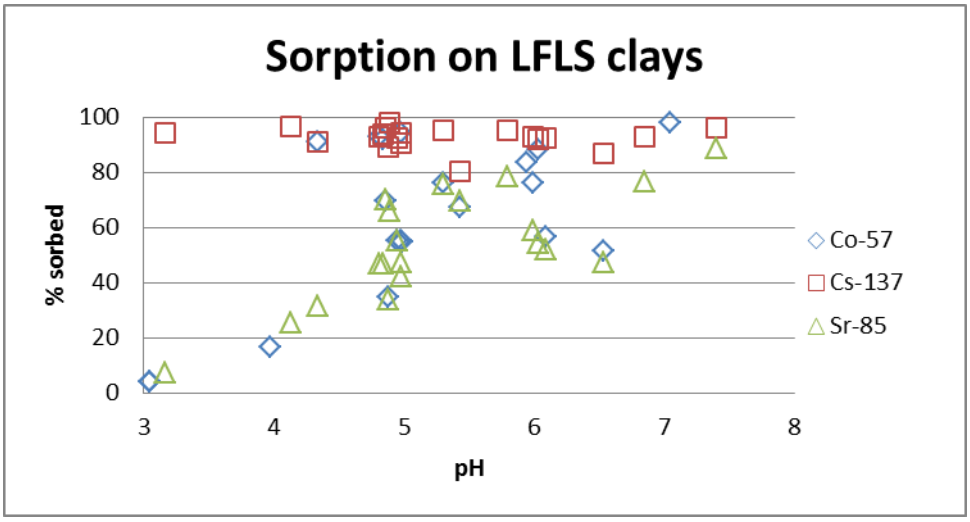


**Figure 5.5: Sorption curves for sorption of Co, Cs and Sr onto LFLS clays CH1A (0.4 – 0.6 m); CH30 (0.4 – 1.0 m), W2D (4.5 – 5.0 m) as a function of pH. Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

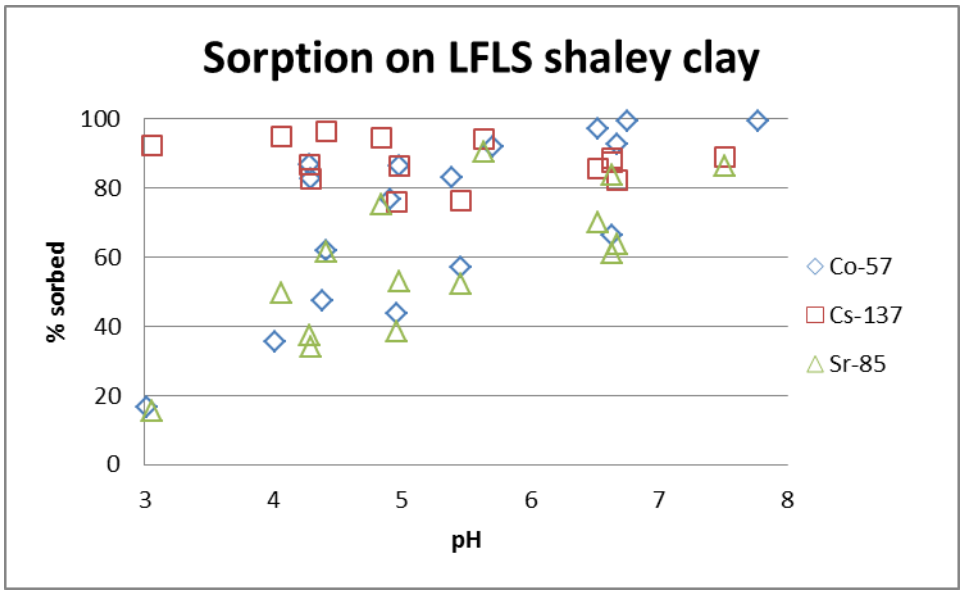


**Figure 5.6: Sorption curves for sorption of Co, Cs and Sr onto LFLS shaley clays CH1A (1.4 – 1.6 m), CH30 (1.0 – 1.2 m) as a function of pH. Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

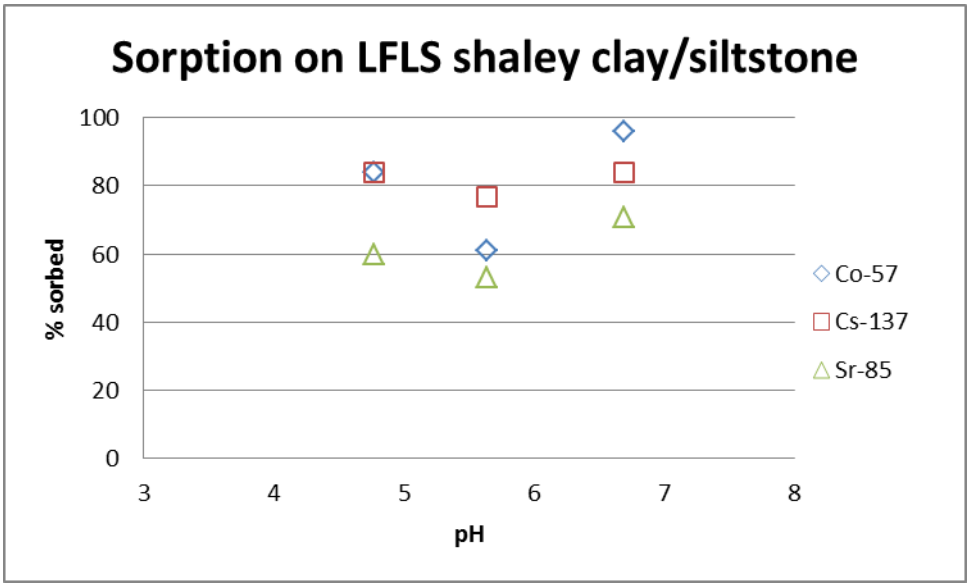


**Figure 5.7: Sorption curves for sorption of Co, Cs and Sr onto LFLS shaley clay/siltstone CH30 (1.4 – 1.6 m) as a function of pH. Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

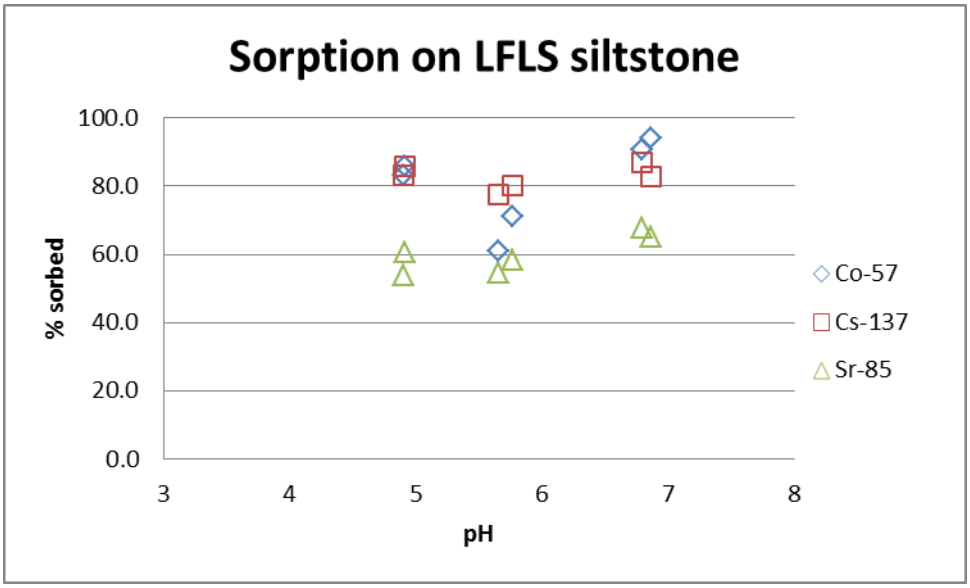


**Figure 5.8: Sorption curves for sorption of Co, Cs and Sr onto LFLS siltstone (1.6 – 1.8 m); as a function of pH. Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

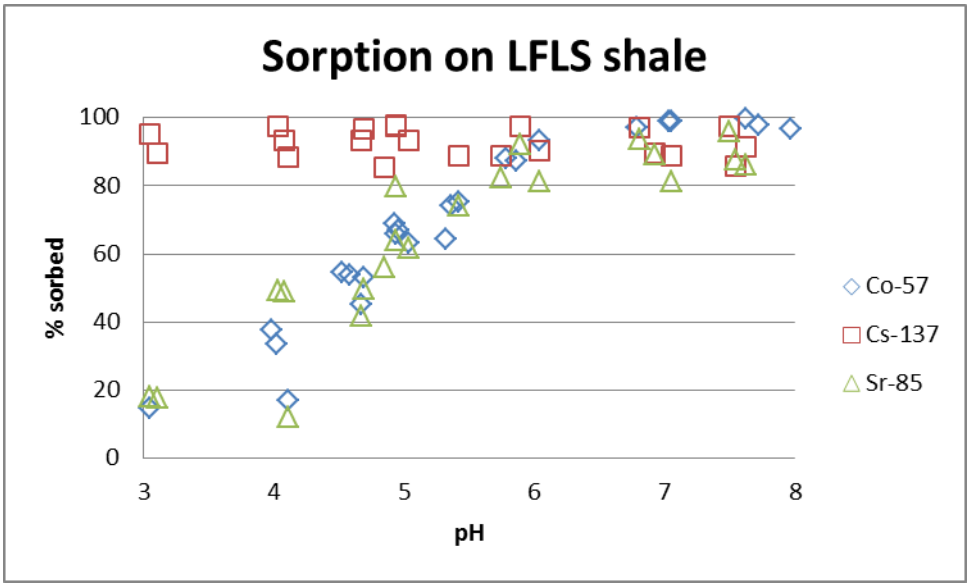


**Figure 5.9: Sorption curves for sorption of Co, Cs and Sr onto LFLS shale CH1A (2.4 – 2.6 m; 3.4 – 3.0 m); W2D (2.5 – 3.0 m) as a function of pH. Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

## 5.4.3. Effect of ionic strength on sorption by CH1A samples.

The effect of ionic strength on sorption was investigated using four samples of CH1A at the various depth intervals and at two ionic strengths, namely 0.01 M and 0.1 M (NaCl). Figure 5.10 shows the sorption curves obtained for Cs, Sr and Co, respectively, as a function of pH for the two ionic strengths. The sorption of Cs ranges from 82% to 98%, with no strong pH or ionic strength dependence.

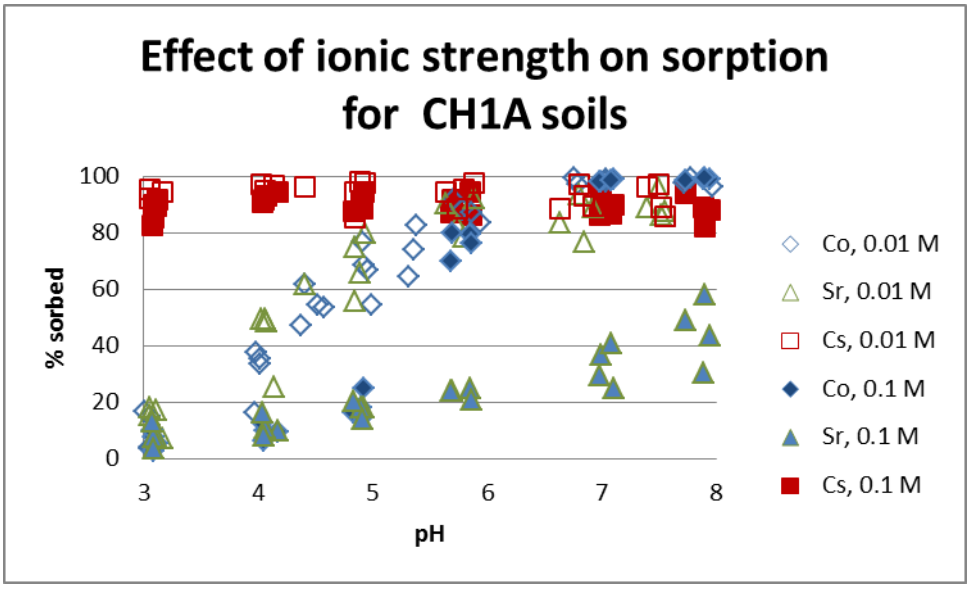


**Fig. 5.10: Sorption of Co, Sr and Co onto CH1A soils (mass loading = 10 g/L; equilibrium with air; 25 ºC) at two ionic strengths. Filled symbols are at the low ionic strength and open symbols are at the high ionic strength.**

However, there is a strong pH and ionic strength dependence for Sr whereby the sorption of Sr is significantly reduced over the entire pH range at the higher ionic strength, with a maximum sorption of < 60%. At pH > 6, maximum sorption (ca. 100%) is obtained at both ionic strengths.

The sorption of Sr at the lower ionic strength shows a greater pH dependency relative to the sorption at higher ionic strength in the pH range 3 to 6. Above pH 6, maximum sorption of ca. 90 to 95% is attained. An ionic strength dependency is also observed for pH values > 4, whereby Sr sorption is significantly reduced at the higher ionic strength. The sorption curves at the higher ionic strength are much shallower in the pH range 3 to 8 with a maximum sorption of < 60% at pH 8.

The sorption of Co is pH dependent at both ionic strengths. At the lower ionic strength, the soil samples have a $pH_{50}$ (i.e. pH of 50% adsorption) of around 4.5. For pH values below 6, the sorption curves are similar for all samples at the higher ionic strength and sorption is typically lower than at the corresponding pH value for the lower ionic strength. The $pH_{50}$ is shifted by *ca*. one pH unit to the right. All materials reach maximum sorption (ca. 100%) above pH 6.5.

# 6. Estimates of Distribution Coefficients ($K_d$) for Co, Cs and Sr at LFLS

The distribution coefficient ($K_d$ value) is the ratio of the radionuclide concentration adsorbed by the solid ($C_s$) and the concentration in the liquid ($C_l$), given by

$$K_d = C_s / C_l \qquad (1)$$

The Kd approach takes no explicit account of sorption mechanisms but assumes that the radionuclide on the solid phase is in equilibrium with the radionuclide in solution and that exchange between these phases is reversible [IAEA 2010].

## 6.1. Estimates of Kd from LFLS sorption data

The $K_d$ values have been estimated from the sorption data in the present work. Compilations of Kd data for the subsamples are provided in Appendix 2.

For the ensuing plots of Kd values vs pH (Figs. 6.1 – 6.8), and the summary tables (Tables 6.1 – 6.3), some assumptions have been made as follows:

- the Kds are deduced from sorption data, typically in the pH range between 4 and 8 so as to be relevant to site conditions [Cendon, 2015].
- Where sorption was > 97.5%, the Kd values are not included in the plots nor for estimates of the mean and standard deviation values (arithmetic and geometric) reported in Tables 6.2 and 6.3. As the amount sorbed relies on a measurement of the equilibrium solution concentration ($C_l$), there is greater uncertainty in this measurement when sorption approaches the maximum value of 100% as the value of $C_l$ would be very low. Hence a conservative uncertainty estimate of 2.5% has been adopted setting an upper bound for absorption at 97.5%, and therefore an upper limit Kd value of ca. 4000 L/kg. Note however, that all Kd estimates are included in the compilation data in Appendix 2.
- A separate Table has been provided for Kd estimates for topsoil/fill due to the topdressing activities that have been undertaken to address subsidence at the site [Payne, 2012]. These may have involved soil materials from beyond the immediate vicinity of LFLS.
- The Tables in the following section provide both the arithmetic mean and geometric mean, and their respective standard deviations. It is recommended that the geometric mean values are used for dose modelling since Kd values in each category (lithologies) will be dependent on a range of factors (e.g. pH, grain size, mineralogy, etc.) and therefore better represented by a geometric mean value.

### 6.1.1. Kd values for Co, Sr and Cs for LFLS materials

Kd values have been estimated for all LFLS soils (Figure 6.1) and for the different lithologies (Figures 6.2 – 6.8).

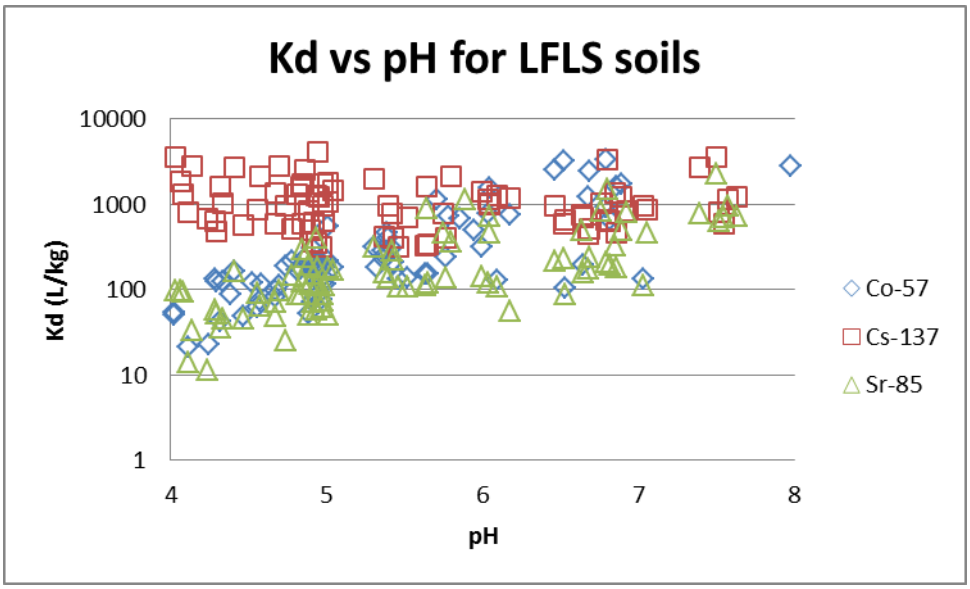


**Figure 6.1: Kd values for Co, Sr and Cs *vs* pH for topsoil and unconsolidated LFLS materials (CH1 (0 – 20 cm), CH1A ( 0.4 – 0.6 m, 1.4 – 1.6 m, 2.4 – 2.6 m, 3.4 – 3.6 m), CH30 ( 0 – 2 m); W2D (0 – 50 cm, 1 – 1.5 m, 2.5 – 3.0 m, 4.5 – 5.0 m). Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

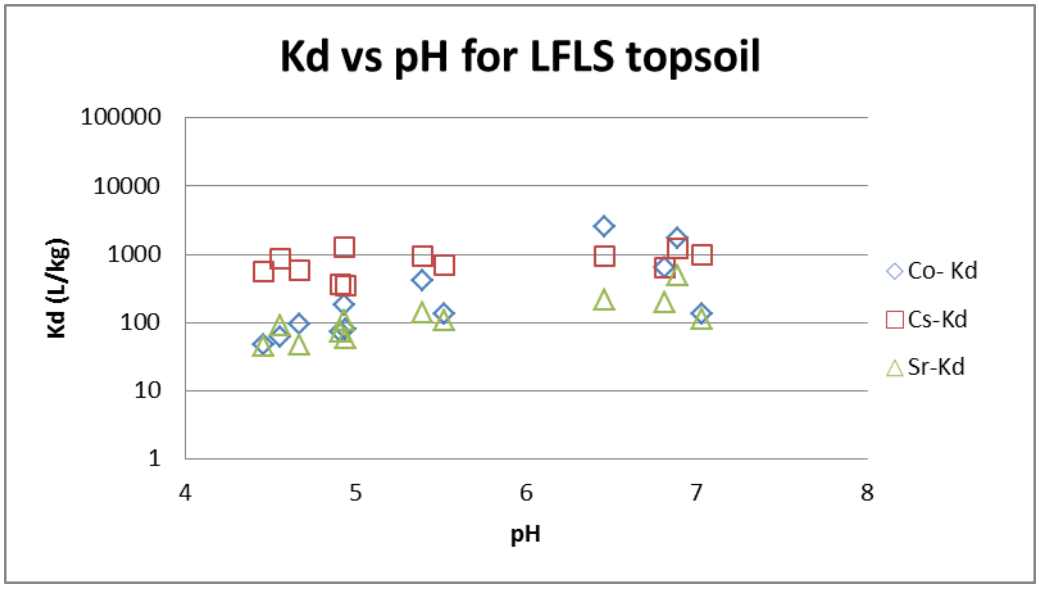


**Figure 6.2: Kd for Co, Sr and Cs *vs* pH for LFLS topsoils CH1 (0 – 0.20 m), CH30 (0 – 0.20 m) and W2D (0 – 0.5 m). Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

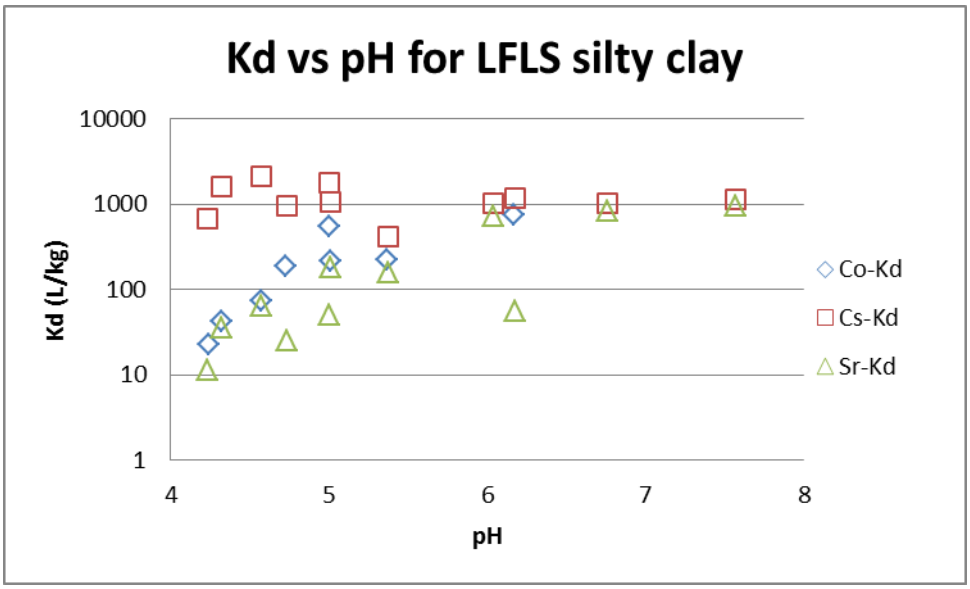


**Figure 6.3: Kd for Co, Sr and Cs *vs* pH for LFLS silty clays CH30 (0.2 – 0.4 m); W2D (1.0 – 1.5 m). Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

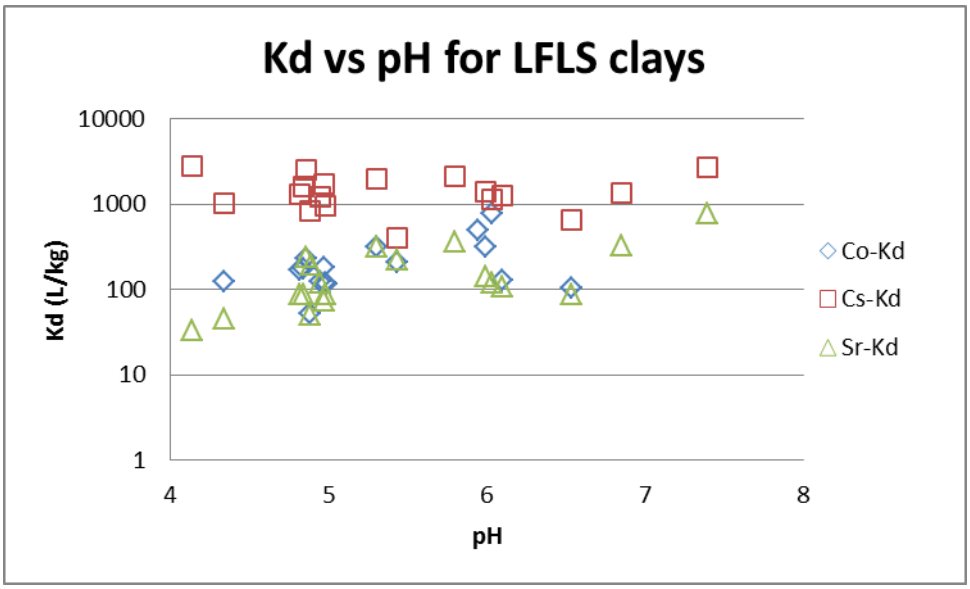


**Figure 6.4: Kd for Co, Sr and Cs *vs* pH for LFLS clays CH1A (0.4 – 0.6 m); CH30 (0.4 – 1.0 m), W2D (4.5 – 5.0 m). Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

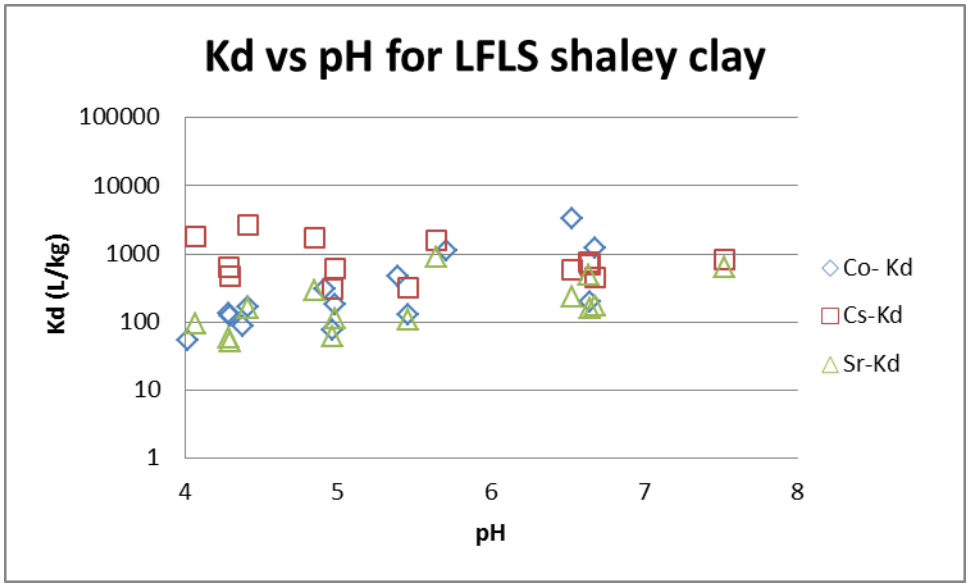


**Figure 6.5: Kd for Co, Sr and Cs *vs* pH for LFLS shaley clays CH1A (1.4 – 1.6 m), CH30 (1.0 – 1.2 m). Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

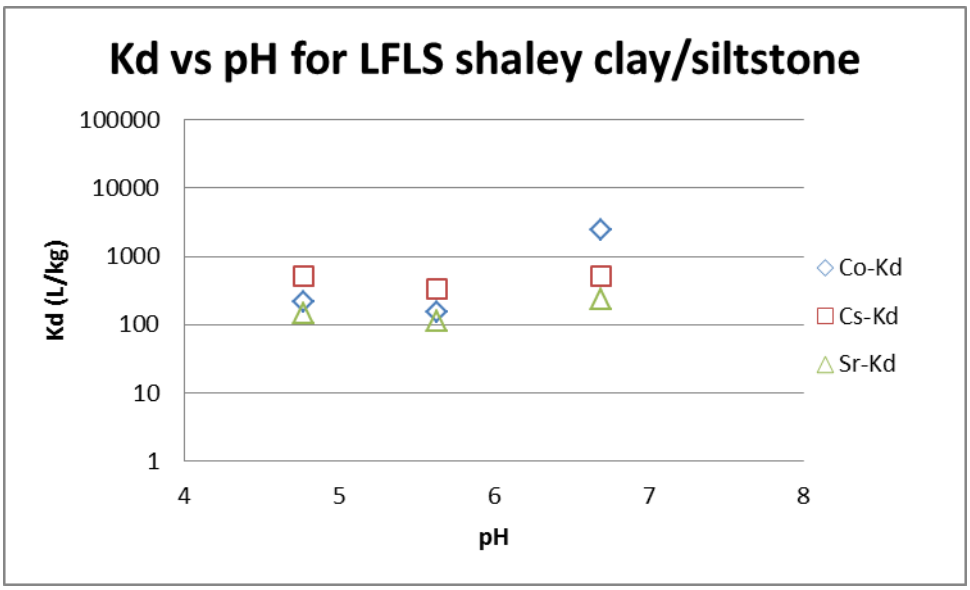


**Figure 6.6: Kd for Co, Sr and Cs *vs* pH for LFLS shaley clay/siltstone CH30 (1.4 – 1.6 m). Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

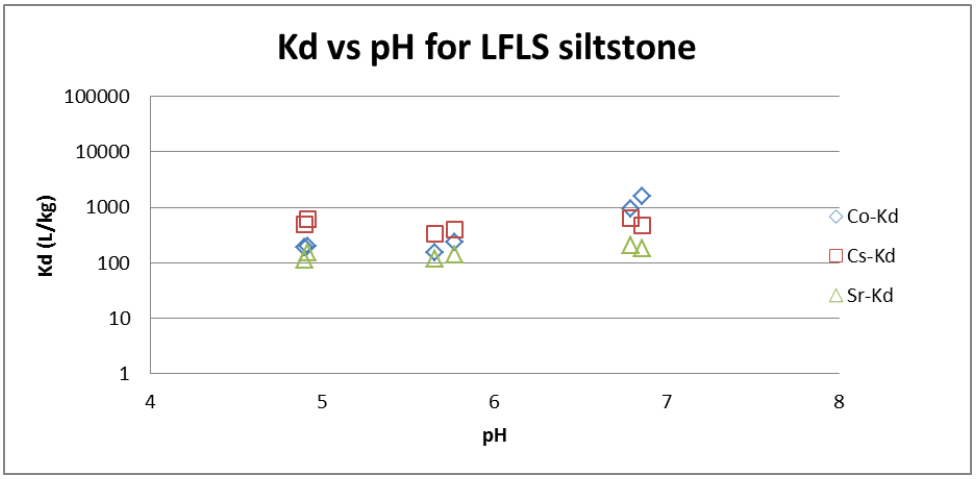


**Figure 6.7: Kd for Co, Cs and Sr *vs* pH for LFLS siltstone (1.6 – 1.8 m); Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

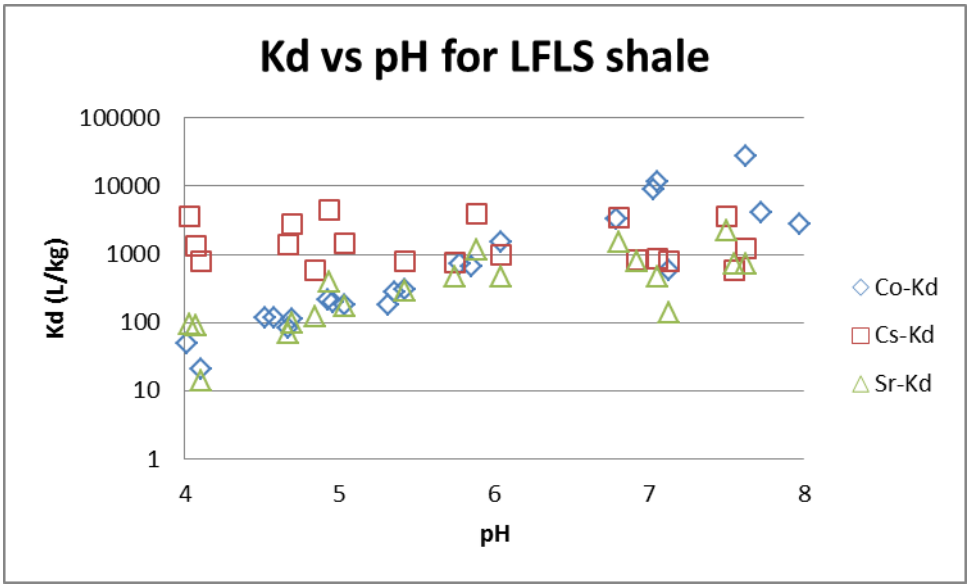


**Figure 6.8: Kd for Co, Cs and Sr *vs* pH for LFLS shale CH1A (2.4 – 2.6 m; 3.4 – 3.0 m); W2D (2.5 – 3.0 m). Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

Tables 6.1 – 6.3 presents a summary of the average Kd values (AM = arithmetic mean and GM =geometric mean) and their respective standard deviations, for all soils and for the various lithological units, as well as the range of Kd values determined (Low = lowest Kd, High =highest Kd), and the number of data points used for the determination (N). The topsoil data has been presented in a separate Table due to the possibility that soils from an external site other than LFLS may have been used in topdressing in the vicinity of the legacy trenches.

The average $K_d$ value for Cs sorption for LFLS topsoil/fill (N = 12) is 730 L/kg with a GSD of 1.5. The average Kd for Co is 210 L/kg (GSD = 3.7) and the average Kd for Sr is 120 L/kg (GSD = 2.0).

**Table 6.1: Kd values for Co, Sr and Cs for LFLS Topsoil/Fill**

| | | | | Kd (L/kg) | | |
|---|---|---|---|---|---|---|
| **LITHOLOGY** | **COREHOLE** | **DEPTH (m)** | | **Co** | **Sr** | **Cs** |
| **TOPSOIL/FILL** | | | **AM** | **510** | **140** | **780** |
| | **CH1** | **0 - 0.2** | **Std dev** | **790** | **120** | **300** |
| | **CH30** | **0 - 0.2** | **LOW** | **48** | **45** | **350** |
| | **W2D** | **0 - 0.5** | **HIGH** | **2500** | **500** | **1300** |
| | | | **GM** | **210** | **110** | **730** |
| | | | **GSD** | **3.7** | **2.0** | **1.5** |
| | | | **N** | **12** | **12** | **12** |

KEY:
AM – arithmetic mean
Std dev – standard deviation of the mean
LOW – lowest Kd value in sample set
HIGH – highest Kd value in sample set
GM – geometric mean
GSD – standard deviation of geometric mean
N – number in sample set
Kds were estimated from batch sorption studies. Ionic strength = 0.01 M, Mass loading of 10 g/L.
Kd values estimated for pH range 4 – 8 for Co, Cs and Sr with average pH of 5.5 (GSD = 1.2).

Excluding the topsoil/fill data, the average $K_d$ value for Cs sorption for all other LFLS soils (N = 66) was 1000 L/kg with a GSD of 1.9. The shaley clay and/or siltstone samples had the lowest average Kd values (440 – 770 L/kg) whereas the average Kd values for Cs for the clay and shale materials were the highest and similar (1300 L/kg, 1400 L/kg, respectively). The average Kd values for Co and Sr (N = 65) were significantly lower than that for Cs for all LFLS soils and similar at 210 L/kg and 180 L/kg. The highest Kds for Co was observed for the shaley clay/siltstone and siltstone materials at 430 L/kg and 360 L/kg, respectively. The highest Kd for Sr was observed for the shale material at 290 L/kg, whereas the Kd for the clay and siltstone materials ranged from 130 – 170 L/kg.

**Table 6.2: Kd values for Co, Sr and Cs for LFLS lithological units**

| | | | | Kd (L/kg) | | |
|---|---|---|---|---|---|---|
| **LITHOLOGY** | **COREHOLE** | **DEPTH (m)** | | **Co** | **Sr** | **Cs** |
| SILTY CLAY | W2D | 1 - 1.5 | AM | 120 | 370 | 1100 |
| | | | Std dev | 97 | 390 | 530 |
| | | | LOW | 23 | 11 | 410 |
| | | | HIGH | 230 | 950 | 2100 |
| | | | GM | 81 | 160 | 1000 |
| | | | GSD | 2.7 | 5.0 | 1.6 |
| | | | N | 5 | 8 | 8 |
| CLAY | | | AM | 210 | 180 | 1500 |
| | CH1A | 0.4 - 0.6 | Std dev | 180 | 170 | 680 |
| | CH30 | 0.4 - 1.0 | LOW | 18 | 33 | 410 |
| | W2D | 4.5 - 5.0 | HIGH | 770 | 780 | 2800 |
| | | | GM | 160 | 130 | 1300 |
| | | | GSD | 2.3 | 2.2 | 1.6 |
| | | | N | 17 | 19 | 18 |
| SHALEY CLAY | | | AM | 540 | 250 | 960 |
| | CH1A | 1.4 - 1.6 | Std dev | 870 | 250 | 700 |
| | CH30 | 1.0 -1.4 | LOW | 53 | 52 | 310 |
| | | | HIGH | 3300 | 890 | 2700 |
| | | | GM | 240 | 170 | 770 |
| | | | GSD | 3.3 | 2.4 | 2.0 |
| | | | N | 14 | 14 | 14 |
| SHALEY CLAY/SILTSTONE | CH30 | 1.4 -1.6 | AM | 940 | 170 | 450 |
| | | | Std dev | 1300 | 66 | 110 |
| | | | LOW | 150 | 110 | 330 |
| | | | HIGH | 2400 | 240 | 520 |
| | | | GM | 430 | 160 | 440 |
| | | | GSD | 4.5 | 1.5 | 1.3 |
| | | | N | 3 | 3 | 3 |
| SILTSTONE | CH30 | 1.6 -2.0 | AM | 550 | 150 | 490 |
| | | | Std dev | 590 | 36 | 120 |
| | | | LOW | 150 | 110 | 230 |
| | | | HIGH | 1600 | 210 | 650 |
| | | | GM | 360 | 150 | 480 |
| | | | GSD | 2.7 | 1.3 | 1.3 |
| | | | N | 6 | 6 | 6 |
| SHALE | | | AM | 590 | 530 | 1700 |
| | | | Std dev | 930 | 580 | 1200 |
| | | | LOW | 21 | 14 | 580 |
| | CH1A | 2.4 - 2.6; 3.4 - 3.6 | HIGH | 3300 | 2300 | 4100 |
| | W2D | 2.5 - 3 | GM | 240 | 290 | 1400 |
| | | | GSD | 3.8 | 3.5 | 2.0 |
| | | | N | 19 | 19 | 17 |
| **ALL SOILS** | | **see above** | **AM** | **450** | **310** | **1300** |
| | **CH1A** | | **Std dev** | **730** | **380** | **890** |
| | **CH30** | | **LOW** | **18** | **11** | **310** |
| | **W2D** | | **HIGH** | **3300** | **2300** | **4100** |
| | | | **GM** | **210** | **180** | **1000** |
| | | | **GSD** | **3.2** | **2.9** | **1.9** |
| | | | **N** | **64** | **69** | **66** |

KEY:
AM – arithmetic mean
Std dev – standard deviation of the mean
LOW – lowest Kd value in sample set
HIGH – highest Kd value in sample set
GM – geometric mean
GSD – standard deviation of geometric mean
N – number in sample set

Kds were estimated from batch sorption studies. Ionic strength = 0.01 M, Mass loading of 10 g/L.
Kd values estimated for pH range 4 – 8 for Co, Cs and Sr with average pH of 5.4 (GSD = 1.2).
Kd values for samples with > 97.5% sorption have not been included in this table. Refer to Appendix for these values.

### 6.1.2. Effect of Ionic strength on Kd values for Co, Sr and Cs for CH1A soils

A comparison of average Kd values for Co, Sr and Cs at two ionic strengths (0.01M and 0.1 M) for CH1A soils is given in Table 6.3. The soils have been classed according to lithology and average Kd values estimated for each lithology as well as average values for all shale, and for all soils combined. For Co, Sr and Cs, the Kd is highest at the lower ionic strength, which suggests that these radionuclides are more mobile at the higher ionic strength. The pH values of the soils range from pH 4 to pH 8 with an average GM of 5.4 (GSD=1.2) at 0.01 M and an average GM of 5.2 (GSD = 1.5) at 0.1 M.

The samples at interval 2.4 – 2.6 m and 3.4 – 3.6 m are both classified as shales yet have very different sorption properties for Cs at the lower ionic strength. The Kd for Cs for the upper layer of shale is significantly lower (780 L/kg) compared to the Kd for Cs for the bottom layer (3500 L/kg). However, this disparity is not observed at the higher ionic strength (730 vs 690 L/kg). The Kd for Sr at the lower ionic strength for the uppermost shale layer is also significantly different to that for the lower layer shale (310 vs 680 L/kg). A possible explanation for this is that the uppermost shale layer has a lower fine clay fraction compared to the other samples as seen in Figure 4.2. The lower shale layer has the highest mica and/or illite component (ca. 11% vs 6 to 8%) which may explain the higher Cs sorption for this layer. The lower layer shale also has a slightly higher iron oxide content compared to the upper layer shale, hence providing more sorption sites.

The average Kd value for Cs at the lower ionic strength is 1500 L/kg and 960 L/kg at the higher ionic strength. The average Kd values for Co and Sr increase with depth at the lower ionic strength, ranging from 100 to 300 L/kg (GM of 230 L/kg) and 230 to 680 L/kg (GM of 350 L/kg), respectively. There is no significant increase observed with depth at the higher ionic strength. The average Kd values for Co and Sr at the higher ionic strength are 42 L/kg and 31 L/kg, respectively.

**Table 6.3: Kd values for Co, Sr and Cs for CH1A lithological units at low and high ionic strengths**

| | | | Kd (L/kg) | | | | | |
|---|---|---|---|---|---|---|---|---|
| CH1A | | | Co | | Sr | | Cs | |
| **LITHOLOGY** | **DEPTH (m)** | | **0.01 M** | **0.1 M** | **0.01 M** | **0.1 M** | **0.01 M** | **0.1 M** |
| CLAY | 0.4 - 0.6 | AM | 210 | 140 | 340 | 43 | 2200 | 1600 |
| | | Std dev | 250 | 210 | 280 | 33 | 650 | 100 |
| | | LOW | 18 | 6 | 33 | 11 | 1300 | 1500 |
| | | HIGH | 490 | 380 | 780 | 94 | 2800 | 1700 |
| | | GM | 100 | 51 | 230 | 33 | 2100 | 1600 |
| | | GSD | 5.2 | 6.3 | 3.3 | 2.3 | 1.4 | 1.1 |
| | | N | 3 | 3 | 5 | 5 | 4 | 5 |
| SHALEY CLAY | 1.4 - 1.6 | AM | 370 | 140 | 430 | 28 | 1600 | 1000 |
| | | Std dev | 400 | 220 | 300 | 12 | 710 | 210 |
| | | LOW | 53 | 11 | 97 | 12 | 760 | 800 |
| | | HIGH | 1100 | 390 | 890 | 43 | 2700 | 1300 |
| | | GM | 220 | 46 | 330 | 26 | 1400 | 1000 |
| | | GSD | 3.1 | 6.7 | 2.3 | 1.6 | 1.6 | 1.2 |
| | | N | 6 | 3 | 6 | 5 | 6 | 5 |
| SHALE | 2.4 - 2.6 | AM | 760 | 87 | 440 | 35 | 820 | 740 |
| | | Std dev | 1300 | 120 | 320 | 23 | 300 | 140 |
| | | LOW | 50 | 14 | 92 | 15 | 580 | 660 |
| | | HIGH | 3300 | 230 | 790 | 78 | 1300 | 980 |
| | | GM | 280 | 40 | 310 | 35 | 780 | 730 |
| | | GSD | 4.4 | 4.6 | 2.7 | 1.7 | 1.4 | 1.2 |
| | | N | 6 | 3 | 5 | 5 | 5 | 5 |
| SHALE | 3.4 - 3.6 | AM | 680 | 110 | 1100 | 51 | 3500 | 720 |
| | | Std dev | 1000 | 180 | 870 | 53 | 130 | 250 |
| | | LOW | 60 | 7 | 96 | 9 | 3400 | 450 |
| | | HIGH | 2800 | 320 | 2300 | 140 | 3600 | 1100 |
| | | GM | 300 | 33 | 680 | 32 | 3500 | 690 |
| | | GSD | 3.9 | 7.4 | 3.6 | 3.0 | 1.0 | 1.4 |
| | | N | 6 | 3 | 5 | 5 | 3 | 5 |
| | | **AM** | 720 | 100 | 760 | 45 | 1800 | 730 |
| **ALL SHALE** | **2.4 - 2.6** | **Std dev** | 1100 | 140 | 700 | 39 | 1400 | 190 |
| | **and** | **LOW** | 50 | 7 | 92 | 9 | 580 | 450 |
| | **3.4 - 3.6** | **HIGH** | 3300 | 390 | 2300 | 140 | 3600 | 1100 |
| | | **GM** | 290 | 37 | 460 | 33 | 1400 | 710 |
| | | **GSD** | 3.9 | 4.9 | 3.2 | 2.3 | 2.3 | 1.3 |
| | | N | 12 | 6 | 10 | 10 | 8 | 10 |
| **ALL SOILS** | **see above** | **AM** | 550 | 120 | 560 | 40 | 1800 | 1000 |
| | | **Std dev** | 870 | 160 | 550 | 32 | 1100 | 420 |
| | | **LOW** | 50 | 7 | 92 | 9 | 580 | 450 |
| | | **HIGH** | 2800 | 390 | 2300 | 140 | 3600 | 1700 |
| | | **GM** | 230 | 42 | 350 | 31 | 1500 | 960 |
| | | **GSD** | 3.7 | 4.8 | 2.9 | 2.1 | 1.9 | 1.5 |
| | | **N** | 21 | 12 | 21 | 20 | 18 | 20 |

KEY:
AM – arithmetic mean
Std dev – standard deviation of the mean
LOW – lowest Kd value in sample set
HIGH – highest Kd value in sample set
GM – geometric mean
GSD – standard deviation of geometric mean
N – number in sample set

Kds were estimated from batch sorption studies. Mass loading of 10 g/L.
Kd values estimated for pH range 4 – 8 for Co, Cs and Sr with average pH of 5.4 (GSD = 1.2) at 0.01 M NaCl and average pH of 5.2 (GSD = 1.5) at 0.1 M NaCl, respectively.
Kd values for samples with > 97.5% sorption have not been included in this table. Refer to Appendix for these values.

## 6.2. Comparison of Kd values with historical site data

There has been only one set of previously reported Kd values for Co, Cs and Sr at the LFLS by Isaac and Mears [1977] in their report titled 'A study of the burial ground used for radioactive waste at the Little Forest Area near Lucas Heights New South Wales'. This report describes laboratory and field work connected with the study of the fate of radionuclides buried at the site [AAEC/E427, 1977]. A description of the samples and some of their work on the cation exchange capacity (CEC) of these materials were described in Section 4.2.2 and Table 4.2 of the present study. The Kd values were determined by batch experiments using borehole waters of different ionic concentrations and pH. A soil/liquid ratio of 0.05 g/L was used and the distribution coefficients were determined on 1 g soil samples (0.25 – 0.5 mm size) after equilibration with three 10 mL portions of filtered borehole water. The soil samples were washed with alcohol, dried at 80 °C and subsequently contacted with 20 mL of filtered borehole water containing the tracer of interest. While Isaac and Mears utilised Co-60 and the present work has utilised Co-57 for the sorption studies, the chemical behaviour of both isotopes will be the same. The borehole water was sourced from two boreholes, representing bore water at low ionic strength (borehole BH10) and bore water at high ionic strength (borehole BHB). The former has a chloride concentration of 1200 mg/L (~0.03 M) and a pH of 7.0 and the latter has a chloride concentration of 3370 mg/L (~0.1 M) and pH of 5.0. The latter is therefore comparable to our experimental conditions with high ionic strength of 0.1 M and average pH of 5.2.

The distribution coefficients of the cations Co-60, Cs-137 and Sr-85 using borehole water is summarised in Table 6.4.

**Table 6.4: Summary of Distribution Coefficients of cations Co-60, Cs-137 and Sr-85 using borehole water in Isaac and Mears study (1977).**

| | Kd (L/kg) | | | | | |
|---|---|---|---|---|---|---|
| | Co-60 | | Cs-137 | | Sr-85 | |
| Soil Fraction | Low IS pH 7.0 | High IS pH 5.0 | Low IS pH 7.0 | High IS pH 5.0 | Low IS pH 7.0 | High IS pH 5.0 |
| F1 | 38 | 9 | 3700 | 2900 | 11 | 1 |
| F2 | 7 | 3 | 2100 | 2100 | 9 | 1 |
| F3 | 5 | 2 | 2500 | 3600 | 7 | 1 |
| F4 | 5 | 2 | 1600 | 2700 | 7 | 1 |

Low IS: BH10 borehole water, pH 7.0. Initial concentrations: $Ca^{2+}$:13 mg/L; $Na^{+}$:840 mg/L; $K^{+}$: 11 mg/L; $Al^{3+}$: < 0.1 mg/L; $Mg^{2+}$: 73 mg/L; $Cl^{-}$: 1200 mg/L
High IS: BHB borehole water, pH 5.0. Initial concentrations: $Ca^{2+}$:170 mg/L; $Na^{+}$:2000 mg/L; $K^{+}$: 24 mg/L; $Al^{3+}$: 0.7 mg/L; $Mg^{2+}$: 320 mg/L; $Cl^{-}$: 3370 mg/L

It is noted that the soil samples used in the study by Isaac and Mears have significantly more kaolinite than any of the samples used in our present study. This may be due to the selective fractionation of the samples by Isaac and Mears whereby their particle size range (0.25 – 0.5 mm) excludes the clay and silt fraction. The methodology and experimental conditions used by Isaac and Mears is also significantly different to that used in our present study so results cannot be directly compared. However, both studies are consistent in showing that the Kd values for Cs are significantly higher than that for Co and Sr.

## 6.3. Comparison of Kd values with IAEA data

The IAEA TRS472 report [IAEA, 2010] presents Kd values grouped according to two criteria, namely the texture/organic matter criterion and the co-factor criterion. For the present study, comparisons of Kd values for Co, Cs and Sr are made according to the texture/organic matter criterion (Table 6.5), a classification on the basis of four main soil groups, namely sand, loam, clay and organic, defined according to the mineral matter (sand and clay mineral percentages) and organic matter content in the soil. Figure 6.9 shows a comparison of Kd values for the LFLS soils with the IAEA TRS472 dataset (Table 6.5) and the historical data set [Isaac and Mears, 1977].

**Table 6.5: IAEA Kd values for Cs, Co and Sr in soils grouped according to the texture/organic matter criterion (L $kg^{-1}$) (reproduced from IAEA TRS472, 2010)**

| Element | Soil Group | N | Mean | GSD* | Minimum | Maximum |
|---|---|---|---|---|---|---|
| **Cs** | All soils | 469 | 1200 | 7.0 | 4.3 | 380 000 |
| | Sand | 114 | 530 | 5.8 | 9.6 | 35 000 |
| | Loam + Clay | 227 | 370 | 3.6 | 39 | 380 000 |
| | Organic | 108 | 270 | 6.8 | 4.3 | 95 000 |
| | | | | | | |
| **Co** | All soils | 118 | 480 | 16 | 2.0 | 100 000 |
| | Sand + loam | 89 | 640 | 16 | 2.0 | 100 000 |
| | Clay | 10 | 3 800 | 5.7 | 540 | 99 000 |
| | Organic | 17 | 87 | 9.5 | 4.0 | 5 800 |
| | | | | | | |
| **Sr** | All soils | 255 | 52 | 5.9 | 0.4 | 6 500 |
| | Sand | 65 | 22 | 6.4 | 0.4 | 2 400 |
| | Loam + clay | 176 | 69 | 5.4 | 2.0 | 6 500 |

*GSD: Geometric standard deviation

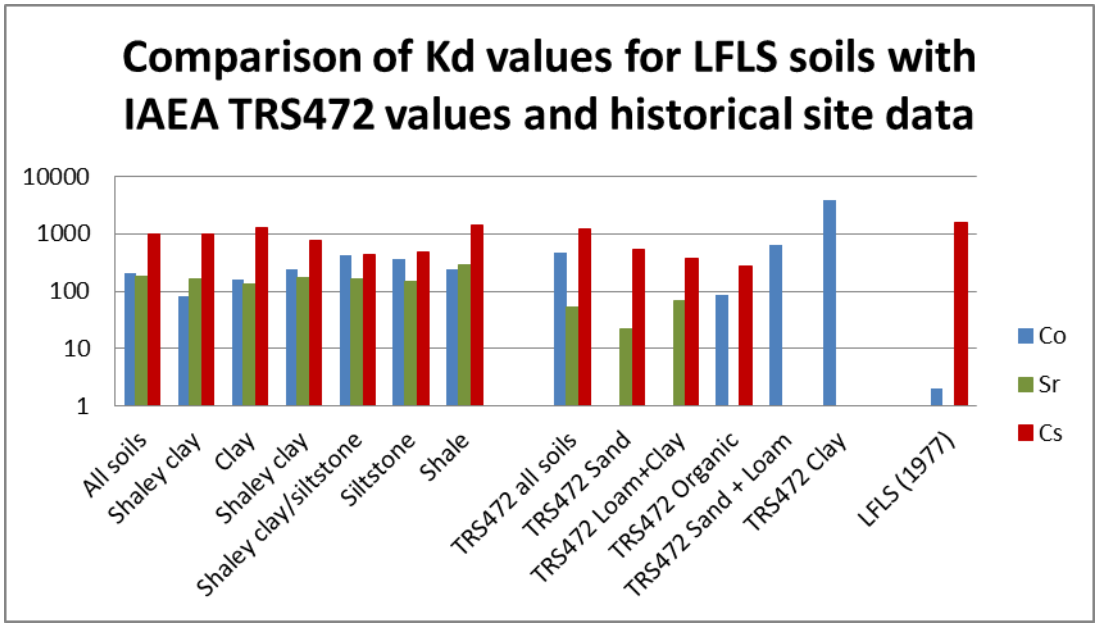


**Figure 6.9: Comparison of Kd values for Co, Cs and Sr for LFLS soil material obtained in the present study with IAEA TRS472 data and LFLS historical data.**

## 6.4. Recommended Kd values for LFLS modelling

Table 6.6 summarises the LFLS site specific Kd values for Co, Cs and Sr recommended for utilisation in various models such as groundwater fate and transport models or dose assessment models. The average Kd values reported in the Table include both the arithmetic mean (AM) and the geometric mean (GM) values from datasets representing the main lithologies at LFLS, excluding the topsoil/fill and deeper samples ( > 5 m W2D samples). The geometric mean indicates the central tendency or typical value of a set of numbers by using the product of their values (as opposed to the arithmetic mean which uses their sum), bearing in mind that the Kds may be dependent on a range of (site) factors such as pH, heterogeneity of samples, particle size distribution, *etc*. It was also noted that where sorption was found to be > 97.5% for the radiotracer, this was not included in the sample set used to arrive at an estimated average Kd value. The justification for this was the anticipated higher (experimental) uncertainties associated with the measurement of very low solution concentrations. Therefore, an upper bound of Kd of *ca* 4000 L/kg was set when determining the average Kd values. However, the maximum Kd value obtained when considering the entire sample set (i.e. including sorption > 97.5%), has been provided in Table 6.6 to allow for sensitivity analysis

**Table 6.6: Recommended dataset of Kd values for Co, Cs and Sr for Dose modelling of LFLS**

| Radionuclide | Estimated average Kd value for LFLS soils (L/kg) | | Range of Kd values based on < 97.5% sorption | Maximum Kd noted* |
|---|---|---|---|---|
| | AM | GM | (L/kg) | (L/kg) |
| **Cs-137** | 1300 | 1000 | Min-max = (310 – 4100) | 4800 |
| **Sr-90** | 310 | 180 | Min – max = (11 – 2300) | 2300 |
| **Co-60** | 450 | 210 | Min-max = (18 – 3300) | 66 000 |

*sorption > 97.5%; AM = arithmetic mean values; GM = geometric mean values

The Kd values were estimated from batch sorption experiments for an ionic strength of 0.01 M (NaCl). The groundwater in the vicinity of the trenches at LFLS is of the Na-Cl type [Cendon et al., 2015]. The pH range considered was pH 4 to 8 with an average pH value of 5.4 ± 1.2. This average pH value is consistent with the reported pH of groundwater across site which varies between 3.9 and 8.0 with an average of 5.4 ± 0.7 [Cendon et al., 2015]. A similar pH range was observed for borewater samples in the study of the LF site by Isaac and Mears (1977). Samples were obtained from three core holes, namely CH30, CH1A, and W2D sourced from various locations at the LFLS site to provide a range of materials representative of the site, both in the vicinity and well away from the legacy trench.

# 7. Sorption of Co on model minerals and simulated soil

## 7.1. Rationale

The purpose of the study was to delineate the key minerals present in LFLS soils that may influence the sorption of Co. A study of Cs and Sr sorption on a simulated LFLS soil and its component model minerals is reported elsewhere [Bots et al., 2019, Bots et al. 2021].

## 7.2. Materials and methods

### 7.2.1. Preparation of model soil

The simulated soil comprised the following mineral components: quartz 45%, kaolinite 25%, illite-smectite 25%, goethite 3%, anatase 2%.

### 7.2.2. Characterisation of model minerals.

BET surface area and CEC determinations were made of the simulated soils and various mineral components as described in Section 4.1.

### 7.2.3. Methodology for sorption studies

Sorption experiments were performed on the component minerals and simulated soils using a Co-57 tracer as per the method outlined in Section 5.2. Kd values were estimated from the sorption curves.

## 7.3. Results and Discussion

### 7.3.1. Geochemical characterisation of model minerals and simulated soil

**Table 7.1: BET surface areas and cation exchange capacities for simulated LFLS soil and component minerals**

| Sample description | BET surface area, m2/g | CEC (cmol $kg^{-1}$) |
|---|---|---|
| Simulated soil | 8.72 | 6.5, 6.4 |
| Illite-smectite | 29.2 | 26.7, 25.5 |
| Goethite | 11.8 | nd |
| Kaolinite | 8.62 | 4.7, 3.7 |
| Anatase | 10.5 | nd |
| Quartz | 0 | nd |

nd = not determined

The measured BET surface area for the simulated soil is similar to the weighted average BET value calculated from the component minerals (10 $m^2g^{-1}$) and is dominated by the illite-smectite component

mineral. The CEC of the simulated soil is significantly lower than that for the illite-smectite component, and comparable to the CEC for the kaolinite component.

### 7.3.2. Sorption of Co on model minerals and simulated soil

The sorption curves for sorption of Co onto the various soil components and the LFLS simulated soil and the respective Kd values are shown in Figures 7.1 and 7.2, respectively.

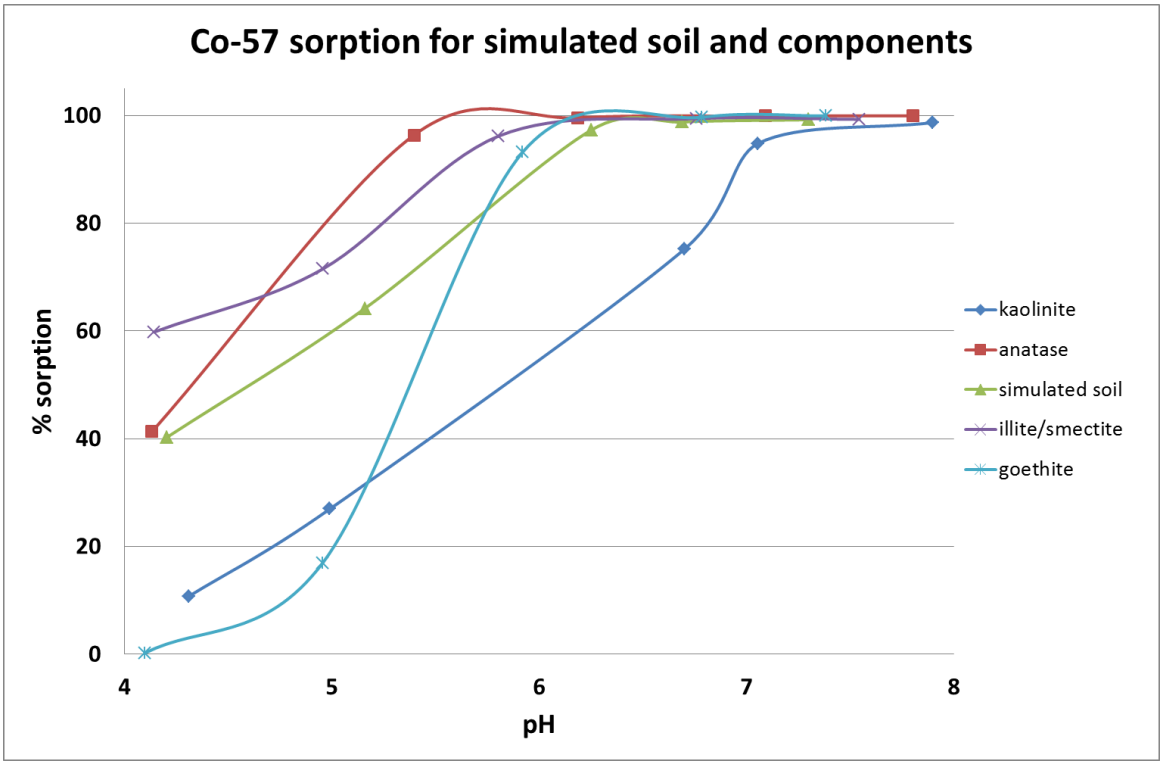


**Figure 7.1: Sorption curves for sorption of Co onto LFLS simulated soil and model mineral components as a function of pH. Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

For the model component minerals, the results show that at *ca*. pH 4, both anatase and illite/smectite are the stronger sorbents relative to goethite and kaolinite, with 40% and 60% sorption of Co, respectively. At this pH, there is less than 10% sorption by kaolinite and no sorption by goethite. At pH 5, all the major soil components show at least 20% sorption, with the maximum sorption of ca. 80% by anatase. Anatase shows *ca*. 100% sorption at pH 5.5, whereas this occurs at pH > 6 for illite/smectite and goethite. For kaolinite, > 95% sorption occurs at pH > 7. The sorption curve of Co for the simulated soil closely mimics the sorption curve for illite/smectite which suggests that the mobility of Co is governed primarily by the illite-smectite phase through surface complexation or ion exchange. However, the small amount of anatase present in the simulated soil may also influence the mobility of Co due to the strong sorption capacity shown by this component.

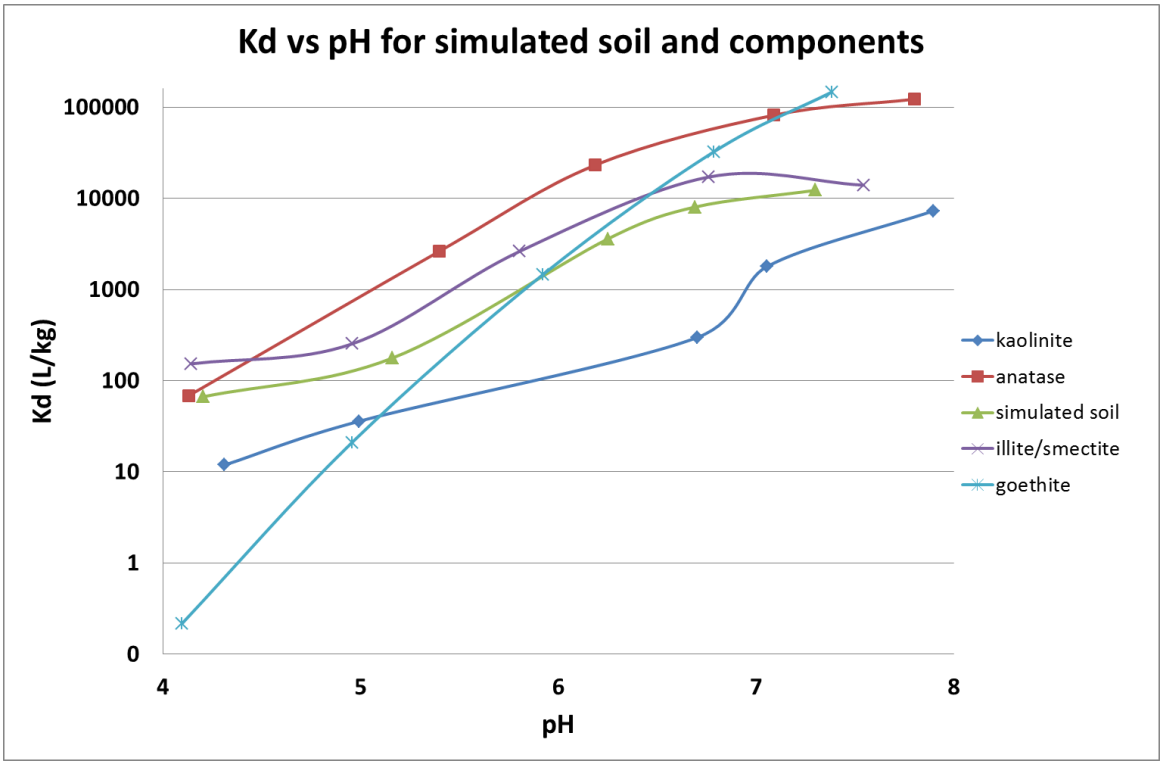


**Figure 7.2: Kd for Co *vs* pH for LFLS model mineral and component minerals. Mass loading of 10 g/L; Ionic strength 0.01 M; air equilibration; T = 25°C.**

# 8. Conclusions and Recommendations

The sorption of Co, Cs and Sr onto mineralogical samples taken from soil cores at the Little Forest Legacy site was investigated for a range of pH values and two ionic strengths. Samples were classed into lithological units, namely topsoil, clay, shaley clay, siltstone and shale.

Strong sorption was observed for Cs over the entire pH range, whereas the sorption of Co and Sr on the soils was found to be pH dependent.

The mobility of Cs is not significantly affected by an increase in ionic strength. The sorption of Sr was significantly reduced at the higher ionic strength over the pH range 4 to 8. The sorption of Co was significantly reduced at the higher ionic strength below pH 6. This has implications for any long term management options which lead to high salinity and/or low pH environments, particularly for the mobility of Co and Sr.

The sorption data was used to produce estimates of distribution coefficient (Kd) values for Co, Sr and Cs under controlled experimental conditions that could be utilised for various site relevant models such as groundwater fate and transport models and/or dose assessment models. Average Kd values for various lithologies and for all soils were determined.

The studies indicate limited mobility of Cs at the LFLS site which is not influenced by changes in pH, nor significantly affected by ionic strength.

At low ionic strength, Kd values for Cs sorption were similar for the majority of soils over the entire pH range, with variations of less than one order of magnitude in Kd values between samples. However, the Kd values for Sr and Co sorption were significantly lower, and varied over several orders of magnitude. Kd values were found to increase with increasing pH values, indicating a decrease in migration rate for Sr and Co with pH.

Model minerals studies for Co sorption indicate that sorption is predominantly controlled by illite/smectite and anatase.

# 9. Acknowledgements

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# Appendix 1: Core Logs of CH1, CH1A, CH30 and W2D

| | | | |
|---|---|---|---|
| **Project ID:** | CES090517-ANS | **Easting:** | 6231951.139 |
| **Project:** | Drilling Investigation | **Northing:** | 313253.421 |
| **Client:** | ANSTO | **Elevation:** | |
| **Location:** | ANSTO - Little Forest Burial Ground | **Environmental Log:** | **CH1** |

CONSULTING EARTH SCIENTISTS
Jones Bay Wharf 19-21, Lower Level Suite 121
26-32 Pirrama Road Pyrmont 2009
PH: (02) 8569 2200 FAX: (02) 9552 4399

| DRILLING INFO. | | | LITHOLOGY | | SAMPLING INFORMATION | | | WELL DETAIL |
|---|---|---|---|---|---|---|---|---|
| Depth | Method | Water | Symbol | Description | Sample ID | Type | FID/PID (ppm) 0 250 500 750 | |
| 0 | | | | TOPSOIL: Silty clay topsoil, brown with rootlets. Moist. | | | | |
| | | | | SILTY CLAY: Brown, moist. Ironstone gravels, trace fine to medium sand and gravels. | | | | |
| 1 | Direct Push | | | CLAY: Brown/orange with red colourations, medium to high plasticity, moist, no odour. Ironstone gravels, trace fine to medium gravels. | | | | |
| | | | | SHALEY CLAY: Orange/brown with ironstone gravels. Dry. | | | | |
| 2 | | | | SHALE: Extremely weathered Shale with siltstone bandings. Light brown and dry. Due to rock formation, sample tube missing from 2 m to 3 m. Push tube refusal @ 3.00 m. | | | | |
| 3 | | | | **End of corehole. Corehole terminated at 3.00 m (practical refusal).** | | | | |
| 4 | | | | | | | | |
| 5 | | | | | | | | |

| | | | |
|---|---|---|---|
| **Drill Company:** | Macquarie Drilling | **Date Commenced:** | 04/08/2009 |
| **Drill Model:** | Geoprobe 7720DT | **Date Completed:** | 04/08/2009 |
| **Hole Diameter (mm):** | 83 | **Logged/checked by:** | M Elbeb |

Sheet: 1 of 1

| | | | |
|---|---|---|---|
| **Project ID:** | CES090517-ANS | **Easting:** | 313253.958 |
| **Project:** | Drilling Investigation | **Northing:** | 6231951.311 |
| **Client:** | ANSTO | **Elevation:** | |
| **Location:** | ANSTO - Little Forest Burial Ground | **Environmental Log:** | **CH1A** |

CONSULTING EARTH SCIENTISTS
Jones Bay Wharf 19-21, Lower Level Suite 121
26-32 Pirrama Road Pyrmont 2009
PH: (02) 8569 2200 FAX: (02) 9552 4399

| DRILLING INFO. | | | LITHOLOGY | | SAMPLING INFORMATION | | | WELL DETAIL |
|---|---|---|---|---|---|---|---|---|
| Depth | Method | Water | Symbol | Description | Sample ID | Type | FID/PID (ppm) 0 250 500 750 | |
| 0 | Direct Push | | | TOPSOIL: Silty clay topsoil, brown with rootlets. Moist. | | | | |
| | | | | SILTY CLAY: Brown, moist. Ironstone gravels, trace fine to medium sand and gravels. | | | | |
| 1 | | | | CLAY: Brown/orange with red colourations, medium to high plasticity, moist, no odour. Ironstone gravels, trace fine to medium gravels. | | | | |
| | | | | SHALEY CLAY: Orange/brown with ironstone gravels. Dry. | | | | |
| 2 | | | | SHALE: Extremely weathered Shale with siltstone bandings. Light brown becoming brown with depth. Dry. Push tube refusal @ 4.00m. | | | | |
| 3 | | | | | | | | |
| 4 | | | | **End of corehole. Corehole terminated at 4.00 m (practical refusal).** | | | | |
| 5 | | | | | | | | |

| | | | |
|---|---|---|---|
| **Drill Company:** | Macquarie Drilling | **Date Commenced:** | 04/08/2009 |
| **Drill Model:** | Geoprobe 7720DT | **Date Completed:** | 04/08/2009 |
| **Hole Diameter (mm):** | 83 | **Logged/checked by:** | M Elbeb |

Sheet: 1 of 1

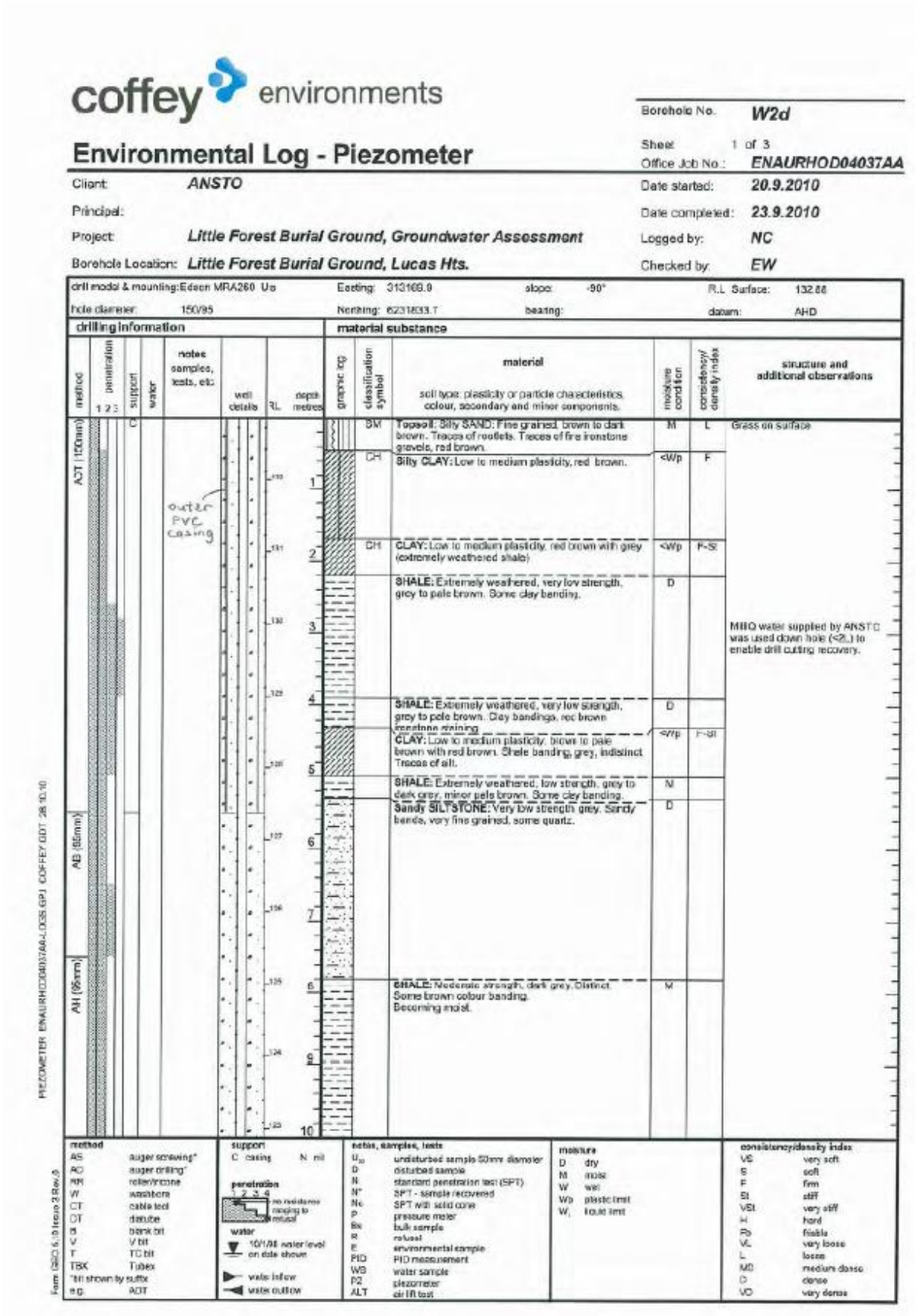
| | | | |
|---|---|---|---|
| **Project ID:** | CES090517-ANS | **Easting:** | 313265.553 |
| **Project:** | Drilling Investigation | **Northing:** | 6231967.183 |
| **Client:** | ANSTO | **Elevation:** | |
| **Location:** | ANSTO - Little Forest Burial Ground | **Environmental Log:** | **CH30** |

CONSULTING EARTH SCIENTISTS
Jones Bay Wharf 19-21, Lower Level Suite 121
26-32 Pirrama Road Pyrmont 2009
PH: (02) 8569 2200 FAX: (02) 9552 4399

| DRILLING INFO. | | | LITHOLOGY | | SAMPLING INFORMATION | | | WELL DETAIL |
|---|---|---|---|---|---|---|---|---|
| Depth | Method | Water | Symbol | Description | Sample ID | Type | FID/PID (ppm) 0 250 500 750 | |
| 0 | Direct Push | | | FILL: Brown, moist. Black charcoal inclusion. Ironstone gravels, trace fine to medium sand and gravels. | | | | |
| | | | | SILTY CLAY: Dark brown, moist. Black charcoal inclusion. Ironstone gravels, trace fine to medium sand and gravels. | | | | |
| 1 | | | | CLAY: Brown/orange with red colourations. Black charcoal inclusions throughout, medium to high plasticity, moist. Ironstone gravels throughout, trace fine to medium gravels. | | | | |
| | | | | SHALEY CLAY: Orange/brown colourations, ironstone gravels, trace fine to medium gravels. Dry. | | | | |
| 2 | | | | SILTSTONE: Extremely weathered siltstone with shale bandings. Light brown/grey becoming brown with depth. Dry. Push tube refusal @ 2.00 m. | | | | |
| | SFA | | | SHALE: Extremely weathered Shale with siltstone bandings. Light brown/grey with depth. Wet from 4.5 m. | | | | |
| 3 | | | | | | | | |
| 4 | | ▼ | | | | | | |
| 5 | | | | End of corehole. Corehole terminated at 5.00 m (limit of investigation). | | | | |

| | | | |
|---|---|---|---|
| **Drill Company:** | Macquarie Drilling | **Date Commenced:** | 05/08/2009 |
| **Drill Model:** | Geoprobe 7720DT | **Date Completed:** | 05/08/2009 |
| **Hole Diameter (mm):** | 83 | **Logged/checked by:** | M Elbeb |

Sheet: 1 of 1



coffey environments

# Environmental Log - Piezometer

Borehole No. W2d
Sheet 1 of 3
Office Job No.: ENAURHOD04037AA
Date started: 20.9.2010
Date completed: 23.9.2010
Logged by: NC
Checked by: EW

Client: ANSTO
Principal:
Project: Little Forest Burial Ground, Groundwater Assessment
Borehole Location: Little Forest Burial Ground, Lucas Hts.

drill model & mounting: Edson MRA260 Ute | Easting: 313169.8 | slope: -90° | R.L. Surface: 132.88
hole diameter: 150/85 | Northing: 6231803.7 | bearing: | datum: AHD

drilling information | material substance

| method | penetration 1 2 3 | support | water | notes samples, tests, etc | well details | RL | depth metres | graphic log | classification symbol | material: soil type: plasticity or particle characteristics, colour, secondary and minor components. | moisture condition | consistency/ density index | structure and additional observations |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| ADT (100mm) | | C | | outer PVC casing | | 132 | 1 | | SM | **Topsoil:** Silty SAND: Fine grained, brown to dark brown. Traces of rootlets. Traces of fine ironstone gravels, red brown. | M | L | Grass on surface |
| | | | | | | 131 | 2 | | CH | **Silty CLAY:** Low to medium plasticity, red brown. | <Wp | F | |
| | | | | | | 130 | 3 | | CH | **CLAY:** Low to medium plasticity, red brown with grey (extremely weathered shale) | <Wp | F-St | |
| | | | | | | 129 | 4 | | | **SHALE:** Extremely weathered, very low strength, grey to pale brown. Some clay banding. | D | | MilliQ water supplied by ANSTO was used down hole (<2L) to enable drill cutting recovery. |
| | | | | | | 128 | 5 | | | **SHALE:** Extremely weathered, very low strength, grey to pale brown. Clay bandings, red brown ironstone staining | D | | |
| | | | | | | 127 | 6 | | | **CLAY:** Low to medium plasticity, brown to pale brown with red brown. Shale banding, grey, indistinct. Traces of silt. | <Wp | F-St | |
| | | | | | | 126 | 7 | | | **SHALE:** Extremely weathered, low strength, grey to dark grey, minor pale brown. Some clay banding. | M | | |
| AB (85mm) | | | | | | | | | | **Sandy SILTSTONE:** Very low strength grey. Sandy bands, very fine grained, some quartz. | D | | |
| AH (95mm) | | | | | | 125 | 8 | | | **SHALE:** Moderate strength, dark grey. Distinct. Some brown colour banding. Becoming moist. | M | | |
| | | | | | | 124 | 9 | | | | | | |
| | | | | | | 123 | 10 | | | | | | |

method: AS auger screwing*; AC auger drilling*; RR roller/tricone; W washbore; CT cable tool; DT diatube; B blank bit; V V bit; T TC bit; TBX Tubex; *bit shown by suffix e.g. ADT

support: C casing; N nil

penetration: 1 2 3 4 (no resistance; rapid to refusal)

water: 10/1/08 water level on date shown; water inflow; water outflow

notes, samples, tests: $U_{50}$ undisturbed sample 50mm diameter; D disturbed sample; N standard penetration test (SPT); N* SPT - sample recovered; Nc SPT with solid cone; P pressure meter; Bk bulk sample; R refusal; E environmental sample; PID PID measurement; WB water sample; PZ piezometer; ALT air lift test

moisture: D dry; M moist; W wet; Wp plastic limit; $W_L$ liquid limit

consistency/density index: VS very soft; S soft; F firm; St stiff; VSt very stiff; H hard; Fb friable; VL very loose; L loose; MD medium dense; D dense; VD very dense

PIEZOMETER ENAURHOD04037AA-L.DGS.GPJ COFFEY.GDT 28/10/10

Form GEO 6.10 Issue 3 Rev 0

coffey environments

# Environmental Log - Piezometer

Borehole No. **W2d**

Sheet 2 of 3

Office Job No.: **ENAURHOD04037AA**

Client: **ANSTO**

Date started: **20.9.2010**

Principal:

Date completed: **23.9.2010**

Project: **Little Forest Burial Ground, Groundwater Assessment**

Logged by: **NC**

Borehole Location: **Little Forest Burial Ground, Lucas Hts.**

Checked by: **EW**

drill model & mounting: Edson MRA260 Ute | Easting: 313169.9 | slope: -90° | R.L. Surface: 132.88

hole diameter: 150/95 | Northing: 6231833.7 | bearing: | datum: AHD

**drilling information** | **material substance**

| method | penetration 1 2 3 | support | water | notes samples, tests, etc | well details | RL | depth metres | graphic log | classification symbol | material: soil type: plasticity or particle characteristics, colour, secondary and minor components. | moisture condition | consistency/ density index | structure and additional observations |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| AH (95mm) | | | ▼ | | | 122 | 11 | | | **SHALE:** Moderate strength, dark grey. Distinct. Some brown colour banding. *(continued)* | M | | |
| | | | | | | | | | | Saturated | W | | |
| | | | | | | 121 | 12 | | | | | | Rapid Groundwater inflow into open borehole. |
| | | | | | | 120 | 13 | | | | M | | |
| | | | | | | 119 | 14 | | | | | | |
| | | | | | | 118 | 15 | | | | | | |
| | | | | | | 117 | 16 | | | **SANDSTONE:** Medium grained, grey to orange. | | | Hawkesbury Sandstone. Difficult to establish moisture due to upper aquifer. |
| | | | | | | 116 | 17 | | | | | | |
| | | | | | | | | | | Orange colouring decreasing, predominantly grey. | | | |
| | | | | | | 115 | 18 | | | | | | |
| | | | | | | 114 | 19 | | | | | | |
| | | | | | | 113 | 20 | | | | | | |

**method**

| | |
|---|---|
| AS | auger screwing* |
| AD | auger drilling* |
| RR | roller/tricone |
| W | washbore |
| CT | cable tool |
| DT | diatube |
| B | blank bit |
| V | V bit |
| T | TC bit |
| TBX | Tubex |

*bit shown by suffix e.g. ADT

**support**: C casing; N nil

**penetration**: 1 2 3 4 — no resistance ranging to refusal

**water**: 10/1/98 water level on date shown; water inflow; water outflow

**notes, samples, tests**

| | |
|---|---|
| $U_{50}$ | undisturbed sample 50mm diameter |
| D | disturbed sample |
| N | standard penetration test (SPT) |
| N* | SPT - sample recovered |
| Nc | SPT with solid cone |
| P | pressure meter |
| Bs | bulk sample |
| R | refusal |
| E | environmental sample |
| PID | PID measurement |
| WS | water sample |
| PZ | piezometer |
| ALT | air lift test |

**moisture**

| | |
|---|---|
| D | dry |
| M | moist |
| W | wet |
| Wp | plastic limit |
| $W_L$ | liquid limit |

**consistency/density index**

| | |
|---|---|
| VS | very soft |
| S | soft |
| F | firm |
| St | stiff |
| VSt | very stiff |
| H | hard |
| Fb | friable |
| VL | very loose |
| L | loose |
| MD | medium dense |
| D | dense |
| VD | very dense |

PIEZOMETER ENAURHOD04037AA-LOGS.GPJ COFFEY.GDT 28.10.10

Form GEO 5.10 Issue 3 Rev.0

# Appendix 2: Compilation of sorption data and Kd data for LFLS soils

**Sorption of Co, Sr and Cs at 0.01 M ionic strength. Experimental conditions 0.01M**

| Corehole | Depth (cm) | Lithology | Target pH | Final pH | % Co-57 sorbed | % Sr-85 sorbed | % Cs-137 sorbed | Kd Co-57 (mL/g) | Kd Sr-85 (mL/g) | Kd Cs-137 (mL/g) |
|---|---|---|---|---|---|---|---|---|---|---|
| CH1 | 0- 20 | Topsoil | 4.5 | 4.67 | 49% | 33% | 86% | 96 | 48 | 595 |
| CH1 | 0- 20 | Topsoil | 5.5 | 5.39 | 80% | 58% | 90% | 406 | 137 | 943 |
| CH1 | 0- 20 | Topsoil | 6.5 | 6.89 | 95% | 83% | 93% | 1725 | 496 | 1235 |
| CH1A | 40-60 | clay | 3 | 3.16 | | 7.2 | 94.4 | | 7 | 1500 |
| CH1A | 40-60 | clay | 4 | 4.13 | | 25.3 | 96.6 | | 33 | 2768 |
| CH1A | 40-60 | clay | 5 | 4.89 | | 66.1 | 98.0 | | 192 | 4819 |
| CH1A | 40-60 | clay | 6 | 5.80 | | 78.8 | 95.5 | | 364 | 2099 |
| CH1A | 40-60 | clay | 7 | 6.85 | | 76.7 | 93.2 | | 323 | 1344 |
| CH1A | 40-60 | clay | 8 | 7.40 | | 88.9 | 96.5 | | 776 | 2662 |
| CH1A | 40-60 | clay | 3 | 3.05 | 4.0 | | | 4 | | |
| CH1A | 40-60 | clay | 4 | 3.98 | 16.4 | | | 18 | | |
| CH1A | 40-60 | clay | 5 | 4.99 | 54.7 | | | 117 | | |
| CH1A | 40-60 | clay | 6 | 5.95 | 83.7 | | | 487 | | |
| CH1A | 40-60 | clay | 7 | 7.04 | 98.1 | | | 5106 | | |
| CH1A | 40-60 | clay | 8 | 8.04 | 97.8 | | | 4312 | | |
| CH1A | 40-60 | clay | 9 | 8.96 | 90.7 | | | 957 | | |
| CH1A | 40-60 | clay | 3 | 3.05 | 3.8 | | | 4 | | |
| CH1A | 140-160 | shaley clay | 3 | 3.06 | | 15.6 | 92.5 | | 18 | 1194 |
| CH1A | 140-160 | shaley clay | 4 | 4.07 | | 49.7 | 94.9 | | 97 | 1835 |
| CH1A | 140-160 | shaley clay | 5 | 4.85 | | 75.1 | 94.6 | | 293 | 1716 |
| CH1A | 140-160 | shaley clay | 6 | 5.64 | | 90.3 | 94.3 | | 891 | 1591 |
| CH1A | 140-160 | shaley clay | 7 | 6.64 | | 83.6 | 88.6 | | 498 | 759 |
| CH1A | 140-160 | shaley clay | 8 | 7.52 | | 86.5 | 89.1 | | 635 | 809 |
| CH1A | 140-160 | shaley clay | 3.0 | 3.01 | 16.6 | | | 18 | | |
| CH1A | 140-160 | shaley clay | 4.0 | 4.02 | 35.6 | | | 53 | | |
| CH1A | 140-160 | shaley clay | 5.0 | 4.91 | 76.6 | | | 307 | | |
| CH1A | 140-160 | shaley clay | 6.0 | 5.70 | 91.9 | | | 1118 | | |
| CH1A | 140-160 | shaley clay | 7.0 | 6.76 | 99.4 | | | 17485 | | |
| CH1A | 140-160 | shaley clay | 8.0 | 7.78 | 99.4 | | | 15772 | | |
| CH1A | 140-160 | shaley clay | 9.0 | 8.88 | 96.0 | | | 2364 | | |
| CH1A | 140-160 | shaley clay | 4.5 | 4.38 | 47.5 | | | 88 | | |
| CH1A | 140-160 | shaley clay | 5.5 | 5.39 | 82.9 | | | 466 | | |
| CH1A | 140-160 | shaley clay | 4.3 | 4.41 | 61.9 | 61.7 | 96.4 | 163 | 162 | 2653 |
| CH1A | 240-260 | shale | 3 | 3.11 | | 17.5 | 89.5 | | 21 | 822 |
| CH1A | 240-260 | shale | 4 | 4.08 | | 48.9 | 93.1 | | 92 | 1308 |
| CH1A | 240-260 | shale | 5 | 4.85 | | 56.1 | 85.5 | | 125 | 579 |
| CH1A | 240-260 | shale | 6 | 5.74 | | 82.6 | 88.7 | | 462 | 769 |
| CH1A | 240-260 | shale | 7 | 6.92 | | 89.1 | 89.7 | | 790 | 838 |
| CH1A | 240-260 | shale | 8 | 7.55 | | 87.8 | 85.8 | | 710 | 594 |
| CH1A | 240-260 | shale | 3 | 3.05 | 14.9 | | | 17 | | |
| CH1A | 240-260 | shale | 4 | 4.02 | 33.4 | | | 50 | | |
| CH1A | 240-260 | shale | 5 | 4.93 | 68.8 | | | 218 | | |
| CH1A | 240-260 | shale | 6 | 5.78 | 88.1 | | | 722 | | |
| CH1A | 240-260 | shale | 7 | 6.79 | 97.1 | | | 3283 | | |
| CH1A | 240-260 | shale | 8 | 7.72 | 97.7 | | | 4157 | | |
| CH1A | 240-260 | shale | 9 | 9.00 | 82.7 | | | 474 | | |
| CH1A | 240-260 | shale | 4.5 | 4.58 | 53.6 | | | 115 | | |
| CH1A | 240-260 | shale | 5.5 | 5.32 | 64.4 | | | 179 | | |
| CH1A | 340-360 | shale | 3 | 3.05 | | 18.2 | 95.3 | | 22 | 2022 |
| CH1A | 340-360 | shale | 4 | 4.03 | | 49.4 | 97.4 | | 96 | 3605 |
| CH1A | 340-360 | shale | 5 | 4.94 | | 79.8 | 97.8 | | 396 | 4534 |
| CH1A | 340-360 | shale | 6 | 5.89 | | 92.1 | 97.6 | | 1145 | 3974 |
| CH1A | 340-360 | shale | 7 | 6.80 | | 93.8 | 97.1 | | 1510 | 3377 |
| CH1A | 340-360 | shale | 8 | 7.50 | | 95.8 | 97.3 | | 2252 | 3582 |
| CH1A | 340-360 | shale | 3 | 2.99 | 10.9 | | | 12 | | |
| CH1A | 340-360 | shale | 4 | 3.98 | 37.8 | | | 60 | | |
| CH1A | 340-360 | shale | 5 | 4.96 | 67.0 | | | 198 | | |
| CH1A | 340-360 | shale | 6 | 5.86 | 87.3 | | | 675 | | |
| CH1A | 340-360 | shale | 7 | 7.03 | 98.9 | | | 8923 | | |
| CH1A | 340-360 | shale | 8 | 7.97 | 96.5 | | | 2774 | | |
| CH1A | 340-360 | shale | 9 | 8.97 | 78.9 | | | 366 | | |
| CH1A | 340-360 | shale | 4.5 | 4.52 | 54.5 | | | 120 | | |
| CH1A | 340-360 | shale | 5.5 | 5.36 | 74.1 | | | 283 | | |

| Corehole | Depth (cm) | Description | Target pH | Final pH | % Co-57 sorbed | % Sr-85 sorbed | % Cs-137 sorbed | Kd Co-57 (mL/g) | Kd Sr-85 (mL/g) | Kd Cs-137 (mL/g) |
|---|---|---|---|---|---|---|---|---|---|---|
| CH30 | 0-20 | Fill | 4.5 | 4.91 | 42% | 42% | 78% | 72 | 72 | 356 |
| CH30 | 0-20 | Fill | 4.5 | 4.56 | 41% | 51% | 91% | 62 | 92 | 852 |
| CH30 | 0-20 | Fill | 5.5 | 7.03 | 57% | 53% | 91% | 134 | 111 | 962 |
| CH30 | 0-20 | Fill | 5.5 | 4.93 | 93% | 53% | 93% | 180 | 110 | 1260 |
| CH30 | 0-20 | Fill | 5.5 | 5.52 | 56% | 50% | 87% | 137 | 107 | 708 |
| CH30 | 0-20 | Fill | 6.5 | 6.46 | 96% | 65% | 89% | 2510 | 213 | 948 |
| CH30 | 20-40 | Silty clay | 4.5 | 4.73 | 66% | 21% | 91% | 188 | 25 | 953 |
| CH30 | 20-40 | Silty clay | 5.5 | 6.17 | 88% | 36% | 92% | 744 | 55 | 1186 |
| CH30 | 20-40 | Silty clay | 5.5 | 5.00 | 95% | 34% | 95% | 546 | 50 | 1750 |
| CH30 | 40-60 | Clay | 4.5 | 4.98 | 55% | 47% | 91% | 121 | 88 | 941 |
| CH30 | 40-60 | Clay | 5.5 | 6.03 | 89% | 55% | 92% | 771 | 119 | 1149 |
| CH30 | 40-60 | Clay | 5.5 | 4.97 | 94% | 42% | 94% | 183 | 72 | 1702 |
| CH30 | 60-80 | Clay | 4.5 | 4.95 | 55% | 55% | 93% | 122 | 121 | 1210 |
| CH30 | 60-80 | Clay | 5.5 | 6.53 | 51% | 47% | 87% | 105 | 89 | 660 |
| CH30 | 60-80 | Clay | 5.5 | 4.84 | 94% | 47% | 94% | 174 | 88 | 1610 |
| CH30 | 80-100 | Clay | 4.5 | 4.34 | 91% | 32% | 91% | 123 | 46 | 1034 |
| CH30 | 80-100 | Clay | 5.5 | 6.09 | 57% | 52% | 93% | 129 | 108 | 1268 |
| CH30 | 80-100 | Clay | 5.5 | 4.81 | 93% | 47% | 93% | 172 | 87 | 1324 |
| CH30 | 80-100 | Clay | 5.5 | 4.88 | 35% | 34% | 90% | 52 | 50 | 841 |
| CH30 | 80-100 | Clay | 6.5 | 5.99 | 76% | 59% | 93% | 318 | 142 | 1376 |
| CH30 | 100-120 | shaley clay | 4.5 | 4.30 | 83% | 34% | 83% | 123 | 52 | 473 |
| CH30 | 100-120 | shaley clay | 5.5 | 6.64 | 66% | 61% | 88% | 195 | 157 | 699 |
| CH30 | 100-120 | shaley clay | 5.5 | 4.98 | 86% | 53% | 86% | 179 | 111 | 622 |
| CH30 | 100-120 | shaley clay | 5.5 | 4.97 | 44% | 39% | 76% | 77 | 62 | 311 |
| CH30 | 100-120 | shaley clay | 6.5 | 6.52 | 97% | 70% | 86% | 3253 | 234 | 591 |
| CH30 | 120-140 | shaley clay | 4.5 | 4.29 | 87% | 37% | 87% | 131 | 58 | 635 |
| CH30 | 120-140 | shaley clay | 5.5 | 5.46 | 57% | 52% | 76% | 131 | 108 | 315 |
| CH30 | 120-140 | shaley clay | 6.5 | 6.68 | 93% | 64% | 82% | 1226 | 173 | 451 |
| CH30 | 140-160 | shaley clay/siltstone | 4.5 | 4.77 | 84% | 60% | 84% | 212 | 148 | 517 |
| CH30 | 140-160 | shaley clay/siltstone | 5.5 | 5.64 | 61% | 53% | 77% | 154 | 112 | 326 |
| CH30 | 140-160 | shaley clay/siltstone | 6.5 | 6.69 | 96% | 71% | 84% | 2447 | 239 | 518 |
| CH30 | 160-180 | siltstone | 4.5 | 4.90 | 83% | 54% | 83% | 190 | 115 | 493 |
| CH30 | 160-180 | siltstone | 5.5 | 5.65 | 61% | 55% | 78% | 153 | 118 | 337 |
| CH30 | 160-180 | siltstone | 6.5 | 6.86 | 94% | 65% | 83% | 1599 | 182 | 469 |
| CH30 | 180-200 | siltstone | 4.5 | 4.91 | 86% | 61% | 86% | 199 | 151 | 600 |
| CH30 | 180-200 | siltstone | 5.5 | 5.77 | 71% | 58% | 80% | 240 | 138 | 399 |
| CH30 | 180-200 | siltstone | 6.5 | 6.79 | 90% | 68% | 87% | 933 | 206 | 648 |

| Corehole | Depth (cm) | Description | Target pH | Final pH | Percentage of Co-57 (region1) in solid phase | Percentage of Sr-85 in solid phase | Percentage of Cs-137 in solid phase | Kd Co-57 (region1) (mL/g) | Kd Sr-85 (mL/g) | Kd Cs-137 (mL/g) |
|---|---|---|---|---|---|---|---|---|---|---|
| W2D | 0- 50 | Topsoil | 4.5 | 4.46 | 33% | 31% | 85% | 48 | 45 | 571 |
| W2D | 0- 50 | Topsoil | 5.5 | 4.94 | 45% | 37% | 78% | 80 | 58 | 353 |
| W2D | 0- 50 | Topsoil | 6.5 | 6.81 | 87% | 66% | 86% | 641 | 194 | 627 |
| W2D | 100-150 | Silty clay | 4 | 4.24 | 18.3 | 9.9 | 87.0 | 23 | 11 | 676 |
| W2D | 100-150 | Silty clay | natural pH | 4.32 | 30.2 | 26.7 | 94.2 | 43 | 36 | 1604 |
| W2D | 100-150 | Silty clay | 5 | 5.01 | 67.7 | 64.1 | 91.2 | 216 | 183 | 1072 |
| W2D | 100-150 | Silty clay | 5.5 | 5.37 | 69.7 | 61.4 | 80.8 | 226 | 156 | 414 |
| W2D | 100-150 | Silty clay | 6 | 6.04 | 98.4 | 87.5 | 91.0 | 6175 | 721 | 1040 |
| W2D | 100-150 | Silty clay | 7 | 6.76 | 99.6 | 89.1 | 91.1 | 24732 | 828 | 1039 |
| W2D | 100-150 | Silty clay | 8 | 7.57 | 99.8 | 90.3 | 91.8 | 65847 | 945 | 1136 |
| W2D | 100-150 | Silty clay | 4.4 | 4.57 | 42.5 | 38.5 | 95.5 | 74 | 63 | 2116 |
| W2D | 250-300 | SHALE | 4 | 4.11 | 16.8 | 11.9 | 88.5 | 21 | 14 | 797 |
| W2D | 250-300 | SHALE | natural pH | 4.68 | 45.1 | 41.6 | 93.2 | 82 | 71 | 1367 |
| W2D | 250-300 | SHALE | natural pH | 4.69 | 53.2 | 49.8 | 96.6 | 113 | 98 | 2827 |
| W2D | 250-300 | SHALE | 5 | 5.04 | 63.1 | 61.8 | 93.3 | 180 | 170 | 1461 |
| W2D | 250-300 | SHALE | 5.5 | 5.42 | 75.1 | 74.2 | 88.7 | 300 | 286 | 781 |
| W2D | 250-300 | SHALE | 6 | 6.04 | 93.4 | 81.3 | 90.1 | 1524 | 467 | 979 |
| W2D | 250-300 | SHALE | 7 | 7.05 | 99.1 | 81.2 | 88.8 | 11382 | 466 | 860 |
| W2D | 250-300 | SHALE | 8 | 7.63 | 99.6 | 86.3 | 91.4 | 27249 | 720 | 1210 |
| W2D | 250-300 | SHALE | 4.8 | 4.94 | 66.0 | 64.0 | 97.5 | 199 | 183 | 4071 |
| W2D | 450-500 | CLAY | natural pH | 4.86 | 69.8 | 70.2 | 96.2 | 231 | 236 | 2523 |
| W2D | 450-500 | CLAY | 5.5 | 5.43 | 67.4 | 69.6 | 80.4 | 205 | 226 | 405 |
| W2D | 450-500 | CLAY | 5 | 5.30 | 76.1 | 76.0 | 95.2 | 326 | 317 | 1979 |
| W2D | 650 | SILTSTONE | natural pH | 7.02 | 91.2 | 51.4 | 69.2 | 1036 | 106 | 225 |
| W2D | 950 | shale | natural pH | 7.13 | 85.6 | 57.8 | 89.0 | 593 | 137 | 807 |
| W2D | 1750 | sandstone | natural pH | 6.93 | 72.7 | 52.8 | 87.2 | 263 | 111 | 675 |

**Sorption of Co, Sr and Cs at 0.1 M ionic strength. Experimental conditions:**

| Corehole | Depth (cm) | Lithology | Target pH | Final pH | % Co-57 sorbed | % Sr-85 sorbed | % Cs-137 sorbed | Kd Co-57 (mL/g) | Kd Sr-85 (mL/g) | Kd Cs-137 (mL/g) |
|---|---|---|---|---|---|---|---|---|---|---|
| KG11A | 40-60 | clay | 3 | 3.1 | 5.7 | 7.5 | 91.7 | 6.0 | 8.1 | 1105 |
| KG11B | 40-60 | clay | 4 | 4.2 | 9.8 | 9.9 | 94.7 | 10.5 | 10.6 | 1746 |
| KG11C | 40-60 | clay | 5 | 4.9 | 24.9 | 18.2 | 94.5 | 32.5 | 21.8 | 1680 |
| KG11D | 40-60 | clay | 6 | 5.8 | 79.4 | 24.9 | 94.6 | 380 | 32.7 | 1711 |
| KG11E | 40-60 | clay | 7 | 7.0 | 98.2 | 37.0 | 93.9 | 5308 | 56.5 | 1493 |
| KG11F | 40-60 | clay | 8 | 7.7 | 98.4 | 49.2 | 94.3 | 6140 | 94.2 | 1597 |
| KG12A | 140-160 | shaley clay | 3 | 3.1 | 7.8 | 7.9 | 90.1 | 8.4 | 8.4 | 899 |
| KG12B | 140-160 | shaley clay | 4 | 4.1 | 9.9 | 11.1 | 93.2 | 10.8 | 12.3 | 1347 |
| KG12C | 140-160 | shaley clay | 5 | 4.9 | 18.4 | 18.4 | 91.6 | 22.3 | 22.2 | 1071 |
| KG12D | 140-160 | shaley clay | 6 | 5.7 | 80.1 | 24.2 | 92.0 | 393 | 31.1 | 1124 |
| KG12E | 140-160 | shaley clay | 7 | 7.1 | 98.9 | 25.2 | 90.2 | 8,769 | 33.1 | 901 |
| KG12F | 140-160 | shaley clay | 8 | 7.9 | 99.2 | 30.4 | 89.0 | 12,070 | 43.0 | 796 |
| KG13A | 240-260 | shale | 3 | 3.1 | 9.9 | 13.3 | 82.8 | 10.9 | 15.1 | 477 |
| KG13B | 240-260 | shale | 4 | 4.0 | 12.8 | 16.5 | 90.9 | 14.3 | 19.4 | 975 |
| KG13C | 240-260 | shale | 5 | 4.8 | 16.4 | 20.5 | 87.5 | 19.6 | 25.8 | 705 |
| KG13D | 240-260 | shale | 6 | 5.7 | 70.1 | 24.2 | 87.1 | 228 | 31.1 | 656 |
| KG13E | 240-260 | shale | 7 | 7.0 | 98.3 | 29.7 | 86.3 | 5480 | 41.2 | 613 |
| KG13F | 240-260 | shale | 8 | 7.9 | 99.0 | 43.9 | 88.0 | 9982 | 77.6 | 729 |
| KG14A | 340-360 | shale | 3 | 3.1 | 2.9 | 3.5 | 85.5 | 2.9 | 3.6 | 580 |
| KG14B | 340-360 | shale | 4 | 4.0 | 6.6 | 8.1 | 91.9 | 7.0 | 8.8 | 1130 |
| KG14C | 340-360 | shale | 5 | 4.9 | 14.6 | 14.3 | 88.8 | 16.5 | 16.1 | 767 |
| KG14D | 340-360 | shale | 6 | 5.9 | 76.3 | 20.9 | 86.5 | 319 | 26.2 | 635 |
| KG14E | 340-360 | shale | 7 | 7.1 | 98.5 | 41.1 | 86.7 | 6145 | 67.4 | 631 |
| KG14F | 340-360 | shale | 8 | 7.9 | 99.3 | 58.3 | 82.2 | 14272 | 136.2 | 450 |

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