MONITORING RADIONUCLIDES IN BEACH DISCHARGES: A NEW PERSPECTIVE ON COASTAL GROUNDWATER RISK NEAR A UK NUCLEAR SITE

BY

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ABSTRACT

Observations of radioactivity from beach springs over a period of four decades provides insights into field scale characteristics of a migrating tritium groundwater plume from the oldest UK nuclear site at Sellafield, in Cumbria. The utility of high resolution spatial and decades long temporal tritium coastal discharge data is demonstrated as an indicator tracer of groundwater flows and radionuclide fate to coastal environmental receptors. Data mined from as far back as 1975 is presented to be of historical significance as one of the earliest unpublished radionuclide contaminant plume investigations in the coastal environment. New field data is presented for tritium activity in beach springs and delineates a distinct asymmetric groundwater plume discharge footprint stretching 2600 metres northwards of the site along the foreshore. The spatial distribution of the tritium is further to the north-west than predicted by the Sellafield Ltd conceptual model and extends beyond the perimeter of the current site groundwater monitoring well network. The spatial footprint was further confirmed by additional sample analysis for technetium-99 with similar contributing source areas. The results of the project suggest that the groundwater flow regime to the north-west of the nuclear site is highly complex and narrow plume contaminant contributions are from more than one source, merging and surfacing in low flow beach springs in the intertidal zone. It is recommended that nuclear sites located near the coast should adopt this simple and inexpensive beach spring monitoring practice as a useful spatial and temporal diagnostic tool at the field scale. This method is complimentary to traditional groundwater monitoring wells and has improved far-field conceptualisation of contaminant sources and resultant pathways to the beach springs and concluded low risk levels to the coastal region (as of 2018).

ACKNOWLEDGEMENTS

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A special acknowledgement to the Groundwater Manager and sampling team, Sellafield Ltd for assistance with fieldwork, provision of Sellafield Ltd groundwater data, borehole logs and GIS maps and more importantly plenty of fun 'geo' field days spent out of the office on the beach. Access to the Sellafield Ltd state-of-the-art groundwater model greatly enhanced the conclusions of this research study and sincere thanks is given to the groundwater modelling lead for carrying out the particle backtracking pathline model runs.

Thanks to my supervisors Mike Rivett, Alan Herbert and John Tellam for all your guidance, encouragement and technical advice over the course of this project. I would also like to thank Gretchel Coldicott for helping navigate all the university systems which has been invaluable to a part-time PGR working at distance.

Finally, I gratefully acknowledge my husband to which this would have not been possible without the ongoing support to help balance a full-time career and research studies, as well as other challenges that life inevitably throws at us. This thesis is dedicated to James.

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CHAPTER 1 INTRODUCTION

1.1 Background

Risk of radionuclide discharge into the coastal environment is a significant issue globally. Nuclear reactors and their associated nuclear waste stores have often been sited near large surface-water bodies for the provision of cooling water during operations, or back-up water supply in an emergency scenario. There are 436 operational nuclear reactors worldwide listed by the IAEA (2017) and my coarse assessment to determine their siting locations indicates nearly half (199) of them are located within proximity to coastal ecosystems (Figure 1.1). Risks to coastal receptors may arise from accidental leakage to ground of radionuclides used in the power generation process (e.g., Gallardo & Marui, 2016; Hirose, 2016; Povinec, 2017; Kaizer et al., 2018) or from legacy sources of by-products and waste from nuclear development and reprocessing activities stored at the surface in specially constructed stores, tanks and ponds or in near surface vaults, trenches or mortuaries (IAEA, 1999). Releases to ground may date back to site inception; for older sites, this may typically be from around the 1950s or even prior.

Nuclear facilities are rigorous in managing inventory of fuel, materials and waste and are conscientious in ensuring routine plant and environmental monitoring is regularly carried out to identify and prevent leaks to ground. There has been continuous improvement in housekeeping over the past few decades supported by implementation of legislation to promote groundwater protection (e.g., Japan Water Pollution Prevention Act 47, 2006; United States Environmental Protection Agency, 1990; The Groundwater Daughter Directive, England 2016; Law of the Peoples Republic of China on Prevention and Control of Water Pollution, 2008; Russia Environmental Policy, 2012) and facilities or practices historically permitted may now be inappropriate. Environmental sampling techniques and laboratory analysis capabilities have also improved and are incorporated into proactive groundwater management site strategies. Such factors, alongside growing source decommissioning activity, may result in leaks to ground nowadays being rarer. Consequently, many radionuclide groundwater plumes monitored may originate from historical sources, and are possibly now in decline (Santschi et al., 1987; Belot et al., 2005; Hughes et al., 2011; Stewart 2012). Proving rates of decline and understanding variability due to rebound effects from later spills, is important in managing risks posed. Mobile radionuclide more easily flushed from sources may offer diagnostic insight into less mobile, attenuated radiological contaminants that more slowly decline (Hu et al., 2008; LiXing et al., 1995). Radionuclides can also be used as applied tracers to understand groundwater flows and recharge (Vogel & Dijken, 1974; Houston, 2007; Wang et al., 2008; Cartwright & Morgenstern, 2012). Insight not only relates to the source term, but also possible receptor impact, for instance pending arrival of more gradual breakthrough and subsequent decline of less mobile, often more toxic, radionuclides.

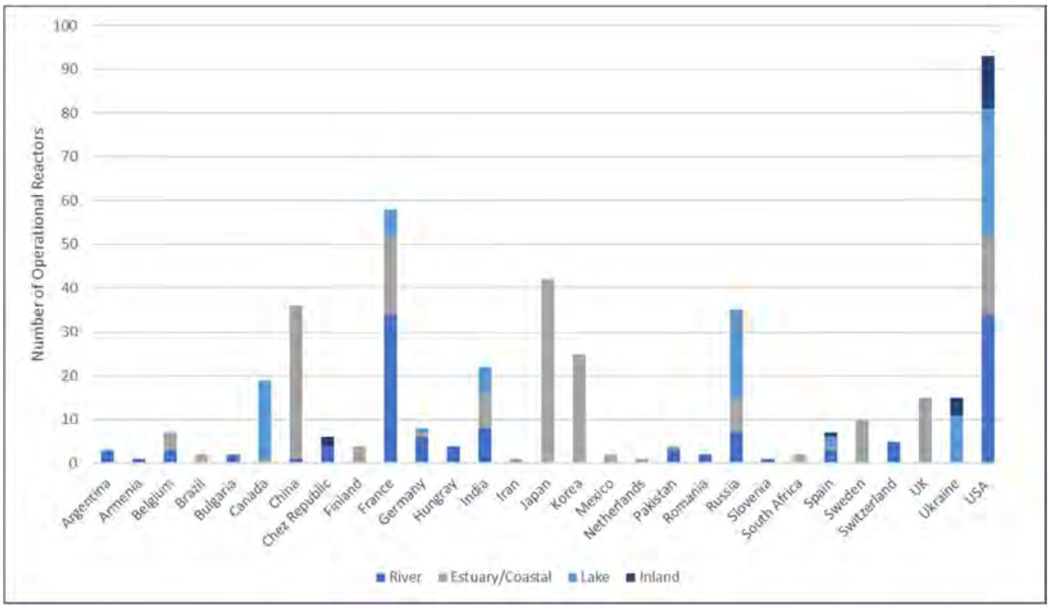


Figure 1-1 Distribution of Siting Locations of Operational Nuclear Reactors Worldwide, coarse assessment from google maps for proximity to surface water bodies (River, Estuary/Coastal, Lake or Inland)

Groundwater transport is significant route that radio-contaminants can enter the biosphere (Dozol & Hagemann, 1993). The principle radionuclides that comprise typical migrating plumes, and which adversely affect groundwater and surface water quality are the most mobile and tend to interact weakly with the geochemical environment with a sufficiently slow decay rate such as tritium, technetium-99, carbon-14, iodine-129 and the less mobile strontium-90 and uranium. Radioactive compounds pose a double threat from both toxicity and damaging radiation to the environment. The migration of radionuclides in groundwater is complex and consequently research studies to evaluate this phenomenon must deal with aqueous chemistry, geochemistry, hydrology, physics and modelling (Dozol & Hagemann, 1993).

Tritium (^{3}H) is a rare radioactive isotope of hydrogen, the nucleus consists of one proton and two neutrons and decays with a half-life of 12.34 years to the stable, inert daughter isotope ^{3}He. Although it is chemically hydrogen, tritium exists within the global environment primarily as part of the water molecule (Plummer et al., 1993). The predominant form of tritiated water (HTO) tends to remain conservatively in water mass and is found throughout the whole hydrosphere (Eyrolle et al., 2018).

The migration of tritium is approximately the flow rate of groundwater, however for other radionuclide contaminants the distribution coefficients of other radioactive nuclides between rock and groundwater are relatively high, and migration rates are slower than tritium. Therefore, tritium can be used as a 'leader' tracer in monitoring of pollution of radioactive nuclides in groundwater, and to predict the migration of following radioactive nuclides and maximum possible area of pollution (Li-Xing et al., 1995). Tritium is thus an ideal tracer for determining timescales for the mixing and the flows of water of the hydrologic cycle, because it is relatively conservative geochemically and has a half-life of

12.34 years it is therefore particularly suited to studying processes that occur of a timescale of less than 100 years (Kendall & Doctor, 2003).

Early application of tritium to hydrologic problems was proposed by Libby and others (Libby, 1953; Kaufman & Libby, 1954; Von Buttler & Libby, 1955; Bergman & Libby, 1957; Eriksson, 1958). The presence of the tritium bomb-pulse from nuclear deterrent testing (1945-1963) was traced through aquifers (Carlston et al., 1960; Allison et al., 1971; Allison & Holmes, 1973) and research then extended to estimating recharge by measuring tritium profiles through the vadose zone (Schmalz & Polzer, 1969; Brendenkemp & Vogel, 1970; Vogel et al., 1974). During the 1980s the bomb-tritium pulse in groundwater began to lose definition. The combined effects of decay and dispersion tended to reduce the tritium concentration levels to values equivalent to that found in precipitation at the time (Phillips & Castro, 2003). The development of the $^{3}H/^{3}HE$ dating method countered this effect (Tolstikhin & Kamensky, 1968) and tritium as a tracer remained relevant. High precision tritium and major ion geochemistry have developed and are now widely used for modern understanding of hydrogeological systems (e.g., Fritz et al, 1996; Le Gal La Salle et al., 2001; Scanlon et al., 2002; Cartwright & Morgenstern, 2012).

There is a legacy of sub-surface radionuclide contamination from nuclear development and power generation over the past 50 years and substantial research is ongoing at some of these nuclear sites because of the extent and complexity of the groundwater plumes. Notable sites with tritium as a leading tracer of contaminated groundwater include Hanford (Fritz et al., 2008), Savannah River (Beals & Hayes, 1995) and the Nevada test site (Hu et al., 2008) in the United States; Glatt Valley in Switzerland (Santschi et al., 1987); Fukushima in Japan (Gallurdo & Marui, 2016) and the largest nuclear site in the UK at Sellafield in Cumbria (Holmes & Hall, 1980). These sites provide examples of radionuclide groundwater contaminants associated with nuclear activities and provide reference sites for future research studies.

1.2 Research Contribution

Tritium (^3H), widely produced in the earlier years of nuclear weapon development and later in commercial electricity power generation, is particularly useful in this regard. Tritium is the dominant radionuclide in releases from nuclear plants (Eyrolle et al., 2018; Vandenhove et al., 2013) and low level radioactive shallow waste disposal sites (Foster et al., 1984; Overcamp.,1982). There is currently no effective technology for removing tritium from nuclear process plant effluents (Jean-Baptiste et al., 2018) so research studies to understand the tritium contribution and behaviour in the environment are highly relevant with the continued investment in worldwide nuclear power generation. When occurring as tritiated water (HTO), it may be simply assumed to migrate with flowing groundwater thereby providing the ideal conservative, non-sorbing, tracer of that fluid motion. Moreover, it has a relatively short half-life of 12.32 years which permits the influence of so-called plume 'natural attenuation' due to radioactive decay to be established. As a mobile but decay-attenuated tracer, it may variously reach receptors of concern. Whilst tritium plumes reaching a coastal receptor is clearly not a desired it is contended that when occurring, maximum use should be made of such data. Understanding tritium's dynamic temporal and heterogeneous spatial footprint of impact at coastal receptors may provide a valuable forensic tool to understanding future fate and risks posed by other radionuclides and improve environmental risk assessments.

The goal of this thesis is to demonstrate the value of the diagnostic far-field beach monitoring by providing new insights into radionuclide contaminant expressions at the coast and explore understanding of pathways to the beach receptor, in order to improve conceptual model understanding and inform of future risks posed. It is hypothesised that a tritium plume migrating from the nearby nuclear site is surfacing locally in the intertidal zone, where the water table depression meets the beach slope and exits onto the shallow foreshore as low flow beach spring features (Sanders, 1998; Horn, 2002; Brassington, 2017). This goal is achieved by a case study at the oldest UK nuclear site at Sellafield, in Cumbria. Sellafield has been operational for over 80 years and was a pioneer for the UK's nuclear industry, it supported national defence, generated electricity for nearly half a century and has developed the ability to safely manage nuclear waste prior to final treatment and storage in a future deep geological disposal facility. The site is home to more than 200 nuclear facilities and the largest inventory of untreated nuclear waste in the world (Sellafield, 2018). The site is actively undergoing decommissioning, a highly complex process extending for the next 100 years. Radiological contaminants found in groundwater at the site include tritium (^{3}H) alongside relatively mobile technetium-99 (^{99}Tc) and strontium-90 (^{90}Sr) (Sellafield Ltd, 2016a), together with other less mobile radionuclides in the unsaturated zone e.g., caesium-137 and actinides. Tritium production has finished at the site and hence on-site sources are assumed to be legacy in origin. This case study is relevant due to the highly complex surface and groundwater interactions in a heterogenous environment and the contaminant plumes have migrated over the last fifty years which will broadly represent a full suite of potential radionuclide contaminants found at most nuclear sites worldwide, and therefore acts as a bounding reference case for all future studies in this research field.

An interdisciplinary approach was used to conduct the research study and considers the interactions between geological and hydrogeological controlling processes, surface water and coastal interactions and prediction of future risks to the environment. The aim and objectives were as follows:

(1) Investigate and collate evidence to propose a preliminary conceptual model (hypothesis) of tritiated groundwater surfacing at beach springs in the intertidal zone, near the Sellafield nuclear site. (Chapter 2)

(2) Obtain new field data characterising the nature and significance of the tritium groundwater plume beach discharge footprint (Chapter 3).

(3) Compare the observed modern discharge footprint to the collation of earlier archived beach spring discharge data spanning some four decades (Chapter 3)

(4) Consider how the observed tritium discharge footprint informs future risks posed from more attenuated radionuclides to the coastal environment (Chapter 4)

(5) Determine the contaminant sources, pathways, and breakthrough characteristics of the tritium groundwater plume at the coastal interface from the understanding developed by the far-field beach springs monitoring technique and refine the Conceptual Model. (Chapter 5).

(6) Consider the wider utility of the approach and make recommendations for future work (Chapter 6)

1.3 Research Design

The approach taken in this study was to focus on the dynamics at the beach- groundwater interface, utilising a tritium contaminant plume as a 'backward' conservative tracer of the groundwater flow processes. Traditional groundwater investigations are source term focused using groundwater monitoring boreholes and predicting pathway and receptor fate by transport and risk modelling. This research study approach is to explore contaminant plumes at the far field receptor perspective at the field scale and adopt a 'backward' looking approach to discover new insights into the understanding of groundwater interactions in the coastal zone. The objectives are met by site specific case study approach drawing on a combination of data mining of historical groundwater investigations and collation of new field data to build a simplified conceptual model.

Figure 1.2 provides an overview of techniques used in this study and cascade of research questioning and gaps that link together the Chapters to develop the beach springs conceptual model. Detailed methodologies can be found in the corresponding empirical chapter, and the field analysis dataset collected is provided in the electronic appendices, any specific data with security or commercially sensitive implications has been omitted.

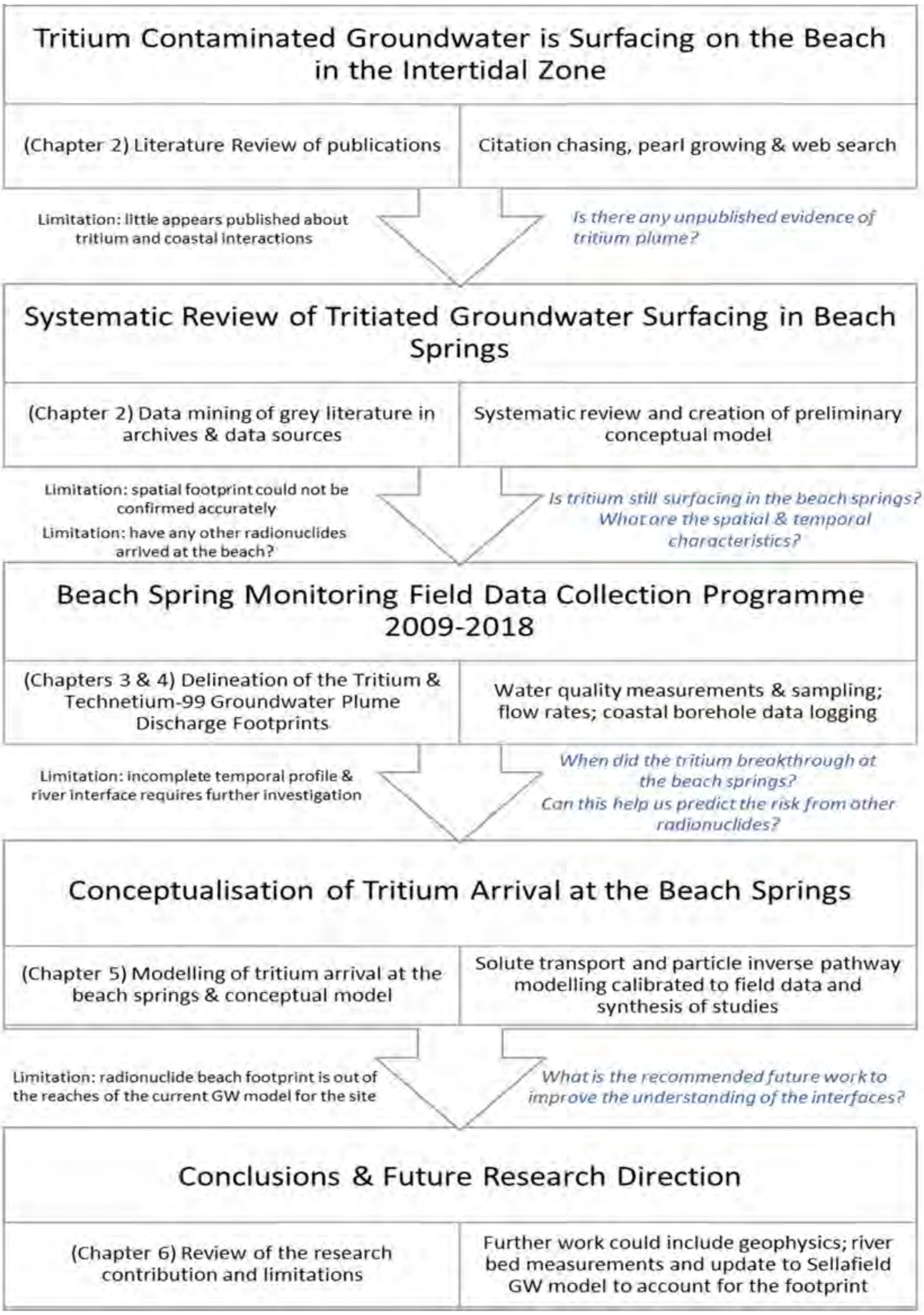


Figure 1-2 Research flow diagram and linkages between the chapters.

1.4 Structure of the Thesis

The thesis is structured around four complementary studies to meet the objectives outlined above and are presented in separate data chapters (Chapter 2-5). Each chapter introduces the aims, describes the applicable literature and methods used, summarises the results with critical discussion and concludes with the contribution to the key thread of the thesis focused around the utility of beach spring monitoring and dynamic interactions.

Chapter 2 collates the output of an extensive desk top investigation into the study area. The aim was to summarise and critically analyse the history of beach investigations 'as told by' the material found in the archives of unpublished reports, data and snippets of miscellaneous information. In the case of Sellafield Site, the continued investment in the groundwater research, monitoring and protection over the last forty years can be traced back to a key 'event' in time when tritium was discovered on the beach in the late 1970s, which triggered further investigations. A preliminary conceptualisation of this groundwater-coastal interface is presented.

Chapter 3 Describes the methods and results of a decade of field investigation to monitor the tritium activity levels in beach spring water samples and presents the evidence for a distinct tritium discharge footprint on the foreshore. Comparisons are drawn with the interpretation of the historical field investigations summarised in Chapter 2.

Chapter 4 Describes the methods and results of a decade of field investigation to monitor the technetium-99 activity levels in beach spring water samples, a radionuclide of concern and presents the evidence for a distinct technetium-99 discharge footprint on the foreshore. Comparisons are drawn with the interpretation of the field investigations summarised in Chapter 3.

Chapter 5 draws together the findings of the previous data chapters to refine the conceptual model understanding of the pathway of the contaminated groundwater to the beach springs and concludes on future risks to the coastal area

Chapter 6 summarises the key conclusions of the beach spring monitoring study to determine radionuclide coastal risk and discusses the wider research implications and makes recommendations for further research direction.

CHAPTER 2 TRITIATED GROUNDWATER SURFACING AT BEACH SPRINGS IN THE INTERTIDAL ZONE NEAR THE SELLAFIELD SITE, UK.

2.1 Introduction

2.1.1 Background

Dynamic groundwater interactions and groundwater contaminant fate at the coastal zone remains a complex multidisciplinary area of research particularly at the field scale. Understanding and quantifying the risk to this receptor from groundwater contaminants is becoming increasingly important due to continued industrialisation and potential for accidental releases of contaminants of anthropogenic origin to groundwater travelling towards coastal zones in particular, there is a research gap in the fate of radionuclides plumes in coastal environments at the field scale. This is a modern concern with the continued reliance and development of worldwide nuclear power capability and the associated environmental risks of release from leaks to ground or nuclear accidents.

Towards the end of the life cycle of nuclear sites there are regulatory requirements for site closure and delicensing. The Scottish Environment Agency, Environment Agency and Natural Resources Wales have produced joint guidance for operators of all UK nuclear sites to manage their radioactive waste, and the condition in which they leave their sites. This Guidance on Requirements for Release from Radioactive Substances Regulation (GRR) requires nuclear operators to produce a waste management plan and a site wide environmental safety case (SWESC) to ensure that the condition of their nuclear site meets the standards for protection of people and the environment, now and into the future (SEPA et al., 2018). In the case where contamination of ground or groundwater arising from the radioactive substance’s activity extends beyond the boundary of the authorised premises, such areas should be considered in the scope of the SWESC. This ensures that all potential sources of exposure to people and impacts on the environment are considered. All nuclear sites with radionuclide groundwater plumes will need to consider the wider implications such as pathways to coastal environmental receptors. These regulations require that historical knowledge and records are preserved for a nuclear operational areas in order to inform flow paths of harmful substances into the environment and this knowledge is required to underpin risk levels for the long term final condition of the site to allow release from regulation, and give confidence in the final fate of the current radionuclide contamination that may be under the nuclear site.

New insights into these coastal pathway interactions are presented by exploring in detail the history of a tritium contaminant plume thought to be surfacing in beach springs in the intertidal zone near the Sellafield nuclear site in the UK, using a systematic review approach. Tritium is a useful groundwater tracer due to its conservative nature and short half-life and was historically produced in large quantities at the Sellafield site. Sellafield was chosen as a site-specific case study, due to a long history of operation (*c*.1940s onwards), known to have complex radionuclide contaminant groundwater plumes and the site is located with proximity (<800m) to the coast. The aim of the systematic review was to gather historical evidence by an extensive search and collation of largely unpublished data relating to potential tritium discharge on the beach prior to the commencement of this research project and implementation of field surveys conducted herein; the systematic review hence covers the period from around 2010 and prior, reaching back to not only the earliest dates of possible plume discharge (*c*.1975), but to nuclear site inception in order to understand possibly tritium source terms contributing.

The objectives of this review were further subdivided in to:

(1) Summarise all knowledge of tritium occurrence in beach springs in coastal zones;

(2) Collate the evidence for tritium contaminated groundwater emerging in beach springs at the Sellafield beach incorporating published and unpublished work from a variety of sources (the vast majority of effort being expended on systematic gathering of unpublished data-archive reports);

(3) Collate the evidence of possible contributing tritium source terms on site, in particular establishing the locations, estimated dates and volumes of release;

(4) Assess and critically evaluate the contribution to the system understanding of groundwater interactions with beach springs from the collated evidence;

(5) Present an interpretation of the evolution of tritium discharges at the Sellafield beach to inform the future Sellafield Site-wide Environmental Safety Case development;

(6) Discuss the gaps and limitations of the review findings and suggested further work.

2.1.2 Research Approach

Tritium was reported to be upwelling in beach springs at the Sellafield beach in 1975 (Hetherington, 1975), following earlier correspondence dating back to a 1973-1974 MAFF report which raised a concern regarding tritium in seawater samples collected around the confluence of the rivers Ehen and Calder and enquiring with the operator whether this was related to a sea burn sewer which triggered a series of detailed investigation works, monitoring in the beach area and liaison between the operator and relevant government agencies. It was important to carry out this preliminary phase of research to unearth any previous investigations, data or scientific reports and analyse previous understanding prior to any further new fieldwork design and commencement. Primary research focus was to gather the relevant records of reporting since or before the discovery of tritium at the beach, as it is common for scientific government organisations that have been operational since the early 1940's to have detailed unpublished internal research/scientific reports stored in national archive facilities and long serving staff from the Sellafield environment department were able to support research direction by giving access to some (incomplete) archive indexes and advice on the types of archive codes/locations to search. In the absence of digital records, it is expected that tacit knowledge over the early decades of operation are lost as personnel retire or departmental reorganisation/closure happens over the years. Another source of data used in this study were water quality samples taken at the beach at regular intervals (quarterly) where the two rivers (Calder and Ehen) converge at the beach which has recorded the presence of tritium historically often referenced as the 'beach confluence' as part of the environmental permit requirements for the site. A systematic review methodology was chosen to firstly understand the extent of the research problem by retrieval and assembly of historical evidence in a structured approach in order to summarise the evolution of the tritium groundwater plume surfacing in the beach springs, which will also have secondary benefits to inform and underpin the future Sellafield site-wide environmental safety case development.

The following sections provide a summary of significant information and data reporting gathered and collated by the systematic review undertaken. Key studies were selected, data was abstracted from the archived reports and collated, the results presented, with a critical analysis of the content and nature of any science investigations that were undertaken in the beach area. All tritium activity results in this Chapter are reported as originally presented in the archived reports and therefore units may change throughout the sections. These values were then converted to Bql^{-1} for the collated results in Sections 2.6 and 2.7 so that results could be meaningfully compared with the new field data presented later in Chapter 3 and is the current standard unit for radioactivity used by the nuclear industry (BIPM, 2019). The conversion factor used was 1 Ci = 3.7×10^{10} Bq. The systematic review was then concluded by reconstructing a timeline of events supported by spatial and temporal data analysis from the individual investigations to draw conclusions about the tritium plume evolution at the Sellafield beach and proposes a preliminary conceptual model of tritium surfacing in beach springs.

2.1.3 Site Specific Study Area

2.1.3.1 Site Setting

The Sellafield site is located on the north-west coast of England, on the edge of the Irish Sea (Figure 2-1). To the east are the Cumbrian Fells and the surrounding land is predominantly open agricultural. The River Ehen lies between the site and the coast, separated by a narrow sand-spit, and flows in a south easterly direction, it joins the River Calder at the south-west of the site where they discharge into the Irish Sea. The Sellafield site is defined by a nuclear site licence boundary and occupies an area of six square kilometres.

Tritium was manufactured at Sellafield between 1958 and 1962 for nuclear deterrent purposes. Cartridges and furnace liners used during the production process were wrapped in plastic and were placed in low level waste disposal trenches and concrete mortuaries on the site, permitted by regulatory guidance at the time (Dunster and Wix, 1959). In 1956 the world's first industrial scale nuclear power station became operational at the Sellafield site. Four reactors produced tritium as a fission by-product of fuel in reactors during electricity generation until their closure in 2003. Solid waste, effluents and liquors containing tritium generated during reprocessing the fuel elements are stored in tank farms and engineered concrete ponds. The location of these tritium sources is in the north and central area of site, where many of the legacy operational buildings reside and is situated approximately 800 to 1700m from the beach-foreshore. Potential mechanisms for accidental tritium release to the environment from the Sellafield site could be to rivers and coastal waters through direct discharge into surface waters, wet or dry deposition by aerial discharges or by direct leaks to ground where tritiated water will migrate with the groundwater with subsequent contaminated groundwater baseflow discharge to rivers or the coastal interface (e.g as beach springs) possible. The latter is the primary pathway of concern in this thesis.

Figure 2-1 : Location map of the beach springs study sampling area from near the Sellafield railway station to the Braystones rail station (marked with a red dots) in west Cumbria. (courtesy of ordnance survey and digimap)

2.1.3.2 Hydrogeological Setting

The geological setting comprises of superficial quaternary drift deposits (unconsolidated silts, sands, clay and gravels) from glacial and fluvio-glacial origin which are variable in thickness and lithology and which are overlain by up to 3 metres of made ground (Figure 2-2). The underlying bedrock is the Triassic Chester Formation, St Bee's Sandstone Member (Ambrose et al., 2014) a fine grained and well cemented sandstone (Michie, 1996; Chaplow, 1996) here, a principal aquifer unit, with bedding planes dipping 23° south-west towards the coast. The inland subsurface is heterogeneous and flow properties can be quite variable over short distances. The depth to bedrock beneath the superficial deposits varies from 3 to 60 metres due to the presence of distinct buried channel features in the bedrock surface. The sandstone surface incorporates a main buried channel which runs westwards across the site (Holmes & Hall, 1980), and is presumed to be likely associated or connected with the ancient path of the River Ehen. There is a north-east trending High Sellafield Fault Zone, this crosses the coastal plain very close to the western site boundary. There is also a series of east to west faults across the site (Smith & Cooper, 2004; Cooper, 2009).

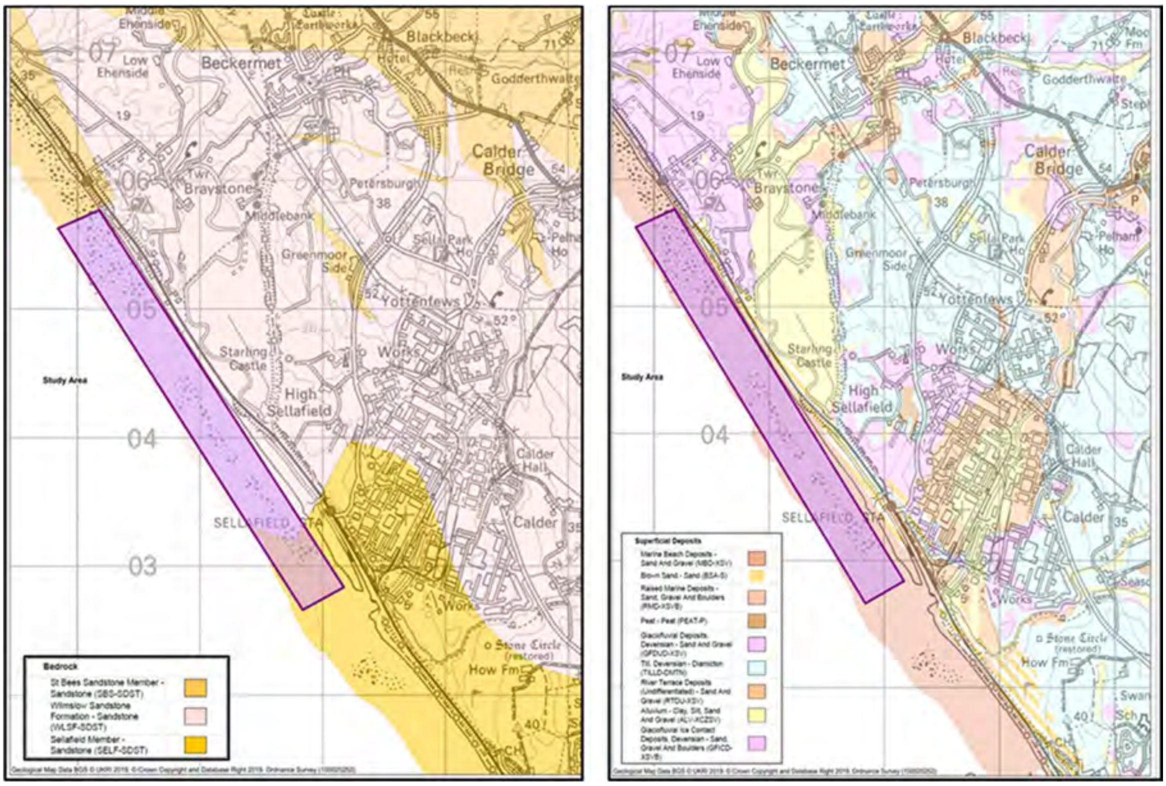


Figure 2-2 Geological setting of the study area bedrock and superficial deposits (Mawby, 2019a; Mawby 2019b) courtesy of digimap.co.uk.

There is hydrological continuity between the drift and sandstones but with complexities in flow directions possibly related to both surface infrastructure at the site and the underlying heterogeneous domain (Black & Brightman, 1996). Broadly, groundwater flows within the drift and sandstone are towards the coast with evidence of base flows to both the River Calder (lower reaches) flowing perpendicular and the River Ehen flowing parallel to the coast. The buried channel, especially where containing more permeable deposits may influence groundwater flow directions with along channel migration of groundwater possible with perhaps some more north-west components of flow. Hydraulic gradients across the site are variable and are defined as approximately 1.0 x 10^{-2} for the drift deposits and 9.5 x 10^{-3} in the sandstone bedrock (Garrick et al., 2010). Typical groundwater travel times to the coast are estimated to be ten to twenty years (Sellafield Ltd., 2016a) however, variations from these groundwater travel time may occur due to the marked heterogeneity in K alongside the potential for predominantly shallow flow pathways as well as initial flow to depth with subsequent discharge at the coast with groundwater surfacing along the saline (wedge) interface. Whilst travel times for tritium will correspond to groundwater (it being a conservative solute), travel times of other radionuclide contaminants such as technetium-99 and strontium-90 may be significantly longer due to their chemical interactions (e.g sorption, ion exchange) with the geological subsurface.

Groundwater flow and associated contaminant plumes from the site may naturally discharge into the lower reaches of the River Calder, the River Ehen or the coastal zone. Average annual environmental monitoring background levels of tritium activity are recorded as 17 Bql^{-1} in seawater and less than 3.5 Bql^{-1} in river waters in this area (Sellafield, 2016b). Known discharges at the coastal foreshore are in the form of relatively low flows from visible beach groundwater springs (Figure 2-3), but also potentially anticipated are deeper, offshore submarine groundwater discharges at the saline interface. A spring discharge zone, often referred to as a 'confluence' in some of the reports (individual spring flows seepages flowing together at the toe of the gravels) was the focus of much historical monitoring. The average tidal range is approximately 9 metres in elevation and 350 metres in lateral distance between mean high and mean low tide and is typical of a flat sand beach gradient found on the Cumbrian coastline.

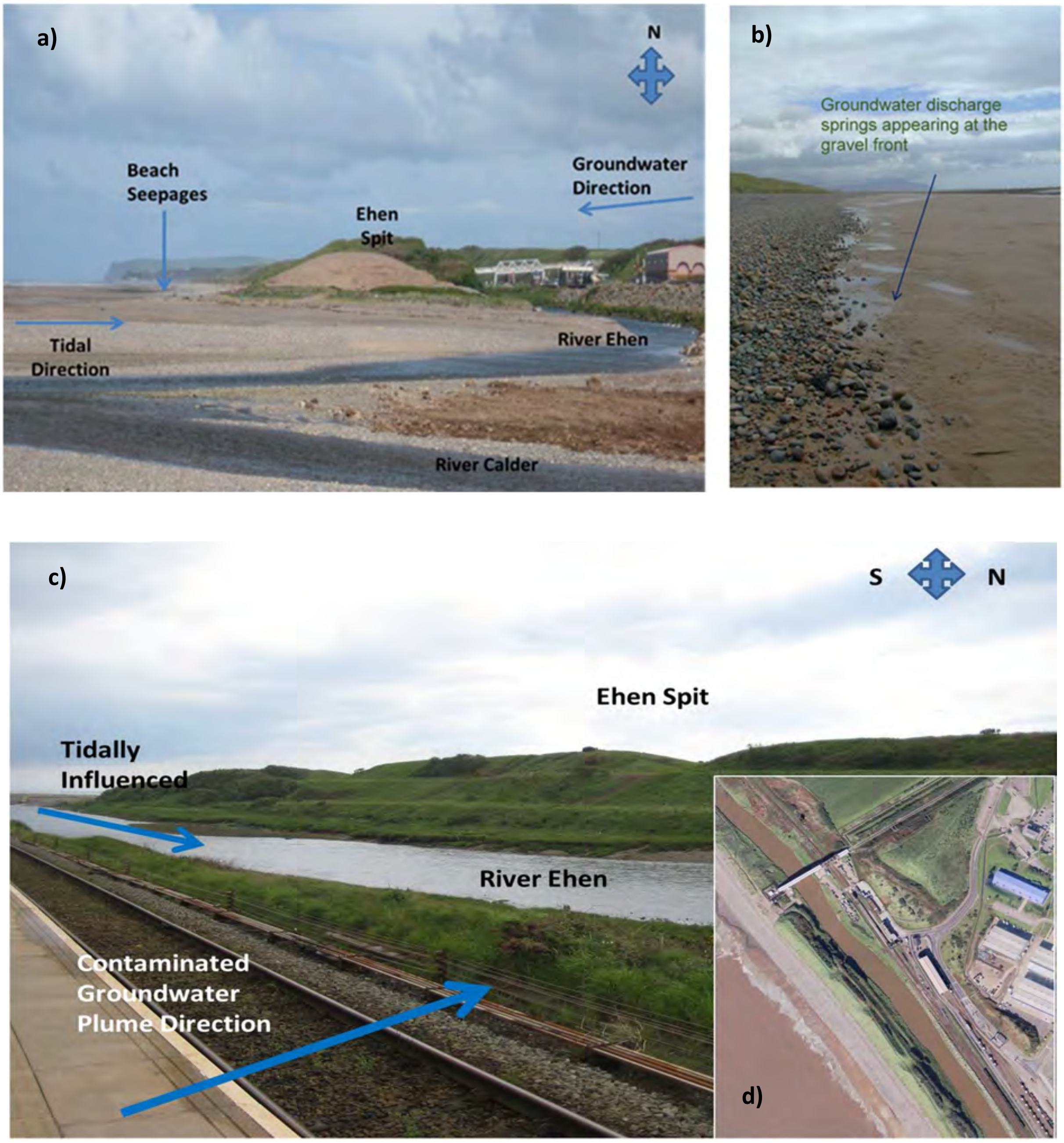


Figure 2-3 Photographs of the study area to show the relationships between a) surface waters and beach seepage discharge areas, b) groundwater springs in the intertidal zone (seepage front), C) river Ehen and the Ehen sand spit, and d) aerial view of site perimeter, river Ehen and intertidal zone and the location of the pipe-bridge.

2.2 Methods and Materials

A literature search was initially performed to determine any peer reviewed publications that related to the research aim both from a broad science basis and specific to the study location and returned very limited results. It was therefore deemed important to consider grey literature searches as part of a thorough systematic review. Grey literature can provide a wide range of information, especially from those who do not publish in academic journals, and advantages include the ability to reduce positive publication bias; have a strong local or regional flavour; present government generated work and may be more detailed than conventional books or articles. The definition of grey literature in the context of this thesis includes access to all previously unpublished reports, discussion papers, working papers, theses and letters (to and from the site) produced by the site owner, as well as dissertations, newsletters, guidelines, government documents, conference papers, posters, technical specifications, regulatory submissions and standards produced by those working within the nuclear, geoscience or appropriate scientific profession. Grey literature is not however bound by the same publishing conventions that characterise 'black literature' and comes in a variety of forms, which poses challenges for data management, extraction and synthesis. Once sources are identified a further stumbling block for inclusion in a review is the assessment of quality, the literature needs to be carefully and systematically examined to judge its trustworthiness, and its relevance and value in the study context (Burls, 2009). To overcome these challenges, it was necessary to develop a study specific quality appraisal approach to collate and critically assess the information relevant to the research question. The method was to carry out a series of steps to complete the review and are summarised in Figure 2-4.

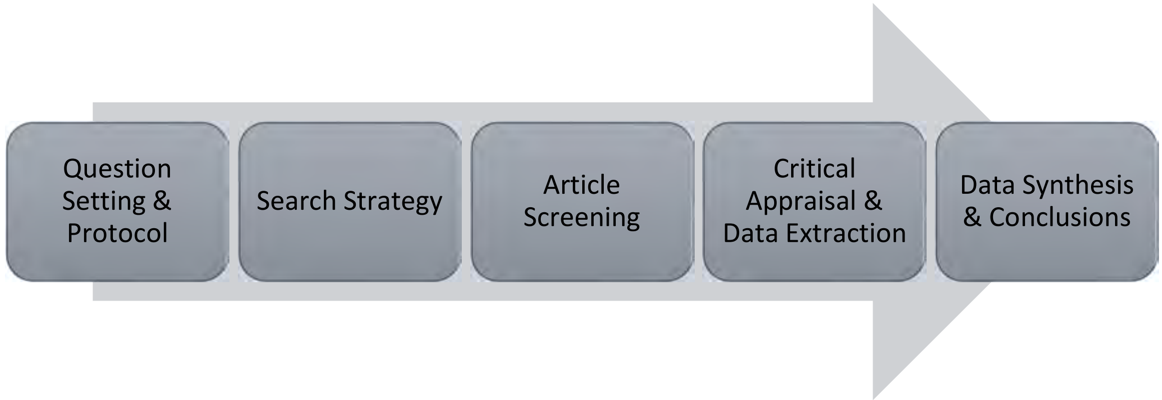


Figure 2-4 Flow chart of systematic review steps

2.2.1 Question setting (aims) and Protocol

The approach is adapted from similar systematic reviews used in both environmental and health studies (Collaboration for Environmental Evidence, 2013; Tyndall, 2010; Moher et al.2009; Green & Higgins,2005). In particular, the AACODS checklist (Tyndall, 2010) provided a good simple starting point and was further tailored to this research question and taken a step further to also apply a quality weighting score in order to assess the importance and reliability. A Microsoft Excel based template was used to capture the assessment against the following criteria: Authority; Accuracy; Coverage; Objectivity; Date and Significance and a set of more specific sub-questions (Table 2-1).

Table 2-1 Categories and sub-questions used to assess the grey literature as part of the systematic review.

Authority	➢ Author associated with reputable organisation. ➢ Professional qualification or a recognised expert? ➢ Cited by others? (grey or published literature) ➢ Higher degree student under expert supervision? ➢ Is it a reputable organisation? (e.g WHO; BGS; govt agency etc.,) ➢ Is there a detailed reference list or bibliography?
Accuracy	➢ Is there a clearly stated aim and is it met? ➢ Is there a stated repeatable methodology? ➢ Is any data collection explicit & appropriate? ➢ Has the document been checked or peer reviewed? ➢ Is it supported by references or credible sources? ➢ Is it representative of work in the field? ➢ Have any laboratory methods been recorded? ➢ Is the minimum detectable level specified (MDL)?
Coverage	➢ Are data inclusions clear? ➢ Are limitations and/or exclusions explicit? ➢ Is the sample size considered meaningful? ➢ Was a range of temporal measurements collected?

	➢ Was a range of spatial measurements collected? ➢ Is the geological/ sub-surface context recorded?
Objectivity	➢ Is the authors rationale for the interpretation clear? ➢ Is the work balanced in presentation? ➢ Is there any identified bias?
Date	➢ Is there a clear date of when the research/report was produced? ➢ Is any data content clearly dated? ➢ Is the bibliography contemporary at the time of writing?
Significance	➢ Is the work related to the research question? ➢ Does it provide context or new knowledge to the research question? ➢ Does it strengthen or refute the current position? ➢ Would the research be lesser without it? ➢ Are the beach springs directly referenced in this report?

2.2.2 Search strategy

The literature search was conducted in two stages. Firstly, a search of published academic databases was completed, followed by citation chasing, pearl growing and web searching. The search was conducted in the English language with no date range limitation. The search syntax was formulated to use 'tritium' and 'beach springs'. 'Seepage' was also substituted to give wider coverage. The search was further extended to include 'radionuclides' and 'groundwater' at 'nuclear plant'. The syntax was split into search clusters which were joined with the Boolean connector 'AND' to give items which made reference in the title, and/or abstract, and/or key words or part of full text. Grey literature searching of the Sellafield Ltd and government records was also included as part of methodology, the research method was to find and follow the 'trail' a snowballing effect by using reports and their references to trace all the internal science reports and field data records related to the subject. Reports, information and field data were gathered from various databases (some now obsolete), as well as finding archived record listings and requesting access from the site owner for boxes of archived non-digital (paper) records that had been put into long term record storage with an offsite data archive company. These boxes were then searched for anything related to the beach springs, tritium in groundwater or Sellafield beach, and then scanned and saved for further analysis. This was a lengthy and iterative process, with the obvious limitation of not being able to categorically declare that all work has been found, there may be unknown gaps, however the records that have been traced appear to align reasonably well over the historical period without obvious significant gaps evident.

2.2.3 Article screening

A first screening of abstracts was conducted, for context and relevance, these were then put into a full document screening, review and critical analysis to further down-select the final studies. The articles/reports found did provide good context, however the main down selection criteria were the direct reference to the beach springs or the area directly surrounding the beach, the River Ehen, groundwater or tritium. The findings from the articles examined against the checklist for relevance and quality assessment are captured in the electronic excel template Appendix 2.1. It should be noted that the Sellafield site has been through a number of changes over the years, it is referred to as the Windscales site in the earlier days, and managed by British Nuclear Fuels Limited (BNFL), now called Sellafield Ltd. Nomenclature may change throughout the reports and summary table but all have been cross checked to ensure the reports are discussing comparable locations.

2.2.4 Critical appraisal and data extraction

The studies are examined for their design and reporting standards and weighted in terms of susceptibility to bias and validity. The reports were ranked as high, medium and low quality against the set scored criteria. There was some duplication of information in the site archive and significant effort was made to trace the original reporting rather than rely on later summarised papers, that often only included key excerpts depending on the audience of the report, without this thorough search important aspects of investigations would not have been gathered particularly those documents such as detailed hydrogeological investigations that were part of a suite of supporting technical references. Appropriate environmental data was then extracted and collated into a data extraction form (excel spreadsheet) and subject to further analysis and presented in the results and discussion (Sections 2.3

to 2.7). It should be noted that any specific nuclear or commercially sensitive data (e.g. inventories) has been omitted from the results sections, however generalised interpretation are still included if relevant.

2.2.5 Data synthesis and conclusions

Each of the individual selected studies are collated and consolidated to form an overall view of the evidence, in this study this was a combination of narrative, quantitative and qualitative information. Gaps and plausible competing explanations of the observed effects are considered, as well as limitations. Key findings, conclusions and recommended future work are summarised at the end of the systematic review.

2.3 Occurrence of Tritium Surfacing in Beach Springs

The primary source of radionuclides in the environment is from anthropogenic nuclear deterrent testing and nuclear fuel cycle processes. Nuclear deterrent tests have contributed to global low-level atmospheric contamination and nuclear site operations often result in high level levels of localised contamination spreading in aquatic pathways (Renshaw et al., 2011). There is published evidence that anthropogenic radionuclides have entered groundwater near nuclear power plants, nuclear deterrent test sites and low-level waste buried in subsurface trenches. Table 2.2 summarises studies of radionuclides in groundwater associated with nuclear facilities. Tritium is found at most of the study locations as it is one of the most prominent radionuclides found in the environment associated with nuclear facilities. The recent most notable published occurrence of tritium in the coastal environment is from the Fukushima nuclear accident (Külahci & Bilico, 2019; Kashiwaya et al., 2017) and proactive management to freeze and treat groundwater is being carried out to manage the environmental risk.

In the late 1960's burial of low-level waste in trenches was common practise and many sites across the United States, Asia, Europe and Australia have recorded tritium plumes migrating in groundwater in terrestrial areas towards rivers and lakes (Zehner, 1983; Foster et al., 1984; Yoon et al., 2010; Hughes et al., 2010). In coastal areas in-land groundwater will typically migrate towards the sea (at lower hydraulic head) and may discharge, overriding the saline wedge interface in the intertidal beach zone as springs or perhaps less obvious (more diffuse) seepages or else offshore as submarine discharges (Cooper, 1959; Glover, 1959; Boufadel, 2000; Abarca et al., 2013; Heiss & Michael, 2014). Review of the published literature surprisingly revealed no field scale studies of radionuclide contaminant plumes surfacing in this zone and no studies have recorded any tritium specifically surfacing in beach springs/seepages, with the exception of Holmes and Hall (1980) who published a paper on the hydrogeology of the Sellafield site, where they made one brief reference to tritium being present in the groundwater, and described the relationship between the groundwater, river Ehen and the beach area. A key publication of geological and hydrogeological studies carried out in the Sellafield area in the 1990s, as part of the NIREX deep geological disposal programme is the special meeting series 29 of the Quarterly Journal of Engineering Geology (Chaplow, 1996).

Table 2-2 Summary of groundwater studies related to anthropogenic radionuclides from worldwide nuclear activities

Site Location	Country	Plant Type	Geology	Environment	Study Interests	Contaminant	Citations
Fukushima	Japan	Nuclear Power Plant	Alluvial deposits, quartenary sand terraces & granites	Coastal	groundwater, atmosphere & ocean inputs from nuclear accident & measures to contain radionuclide contaminants	^{3}H, ^{90}Sr, ^{137}Cs-137, ^{129}I, ^{14}C	Kulahci & Bilici, 2019; Kashiwaya et al., 2017; Gallardo & Marui, 2016; Hirose, 2016; Kaizer et al.,2018; Povinec et al., 2017;
Semi Palantisk	Kazakhistan	Nuclear Test Site (the Polygon)	quarternary sediments	Rivers and Atomic Lakes	groundwater contamination and speciation in well waters, streams and lakes	^{238}U, $^{239\text{-}240}Pu$, ^{241}Am, ^{3}H, ^{137}Cs, ^{90}Sr	Malkovsky et al., 2009
Eurex Plant, Saluggia. Vercelli	Italy	Spent Fuel Reprocessing	Quarternary glacial & fluvial depositis of gravel, sand & silts	River	hydrogeological studies/conceptual model	^{90}Sr, ^{137}Cs, ^{241}Am, $^{239/240}Pu$	Lezzi et al., 2009
Naa-ri, Korean Penisula	Korea	Radioactive repository site	Paleozoic sedimentary rocks	mountain range/streams catchment	monitoring of surface waters and groundwaters using 3H as a leader radionuclide and 14C	^{3}H, ^{14}C	Yoon et al, 2010
Andreeva Bay Shore, Technical Base	NW Russia	spent fuel & radioactive waste facilities	marine (sands) & glacial (conglomerate moraines) sediments	terrestrial & marine	studies of soil and groundwater contamination	^{90}Sr, ^{137}Cs	Standing et al.,2009
VLLW disposal site. Long-men Mountain	China	Candidate Disposal Site	quarternary & siluarian alluvial deposits (sands & gravels)	Confluence of 2 Rivers	geological/hydrogeological studies to calibrate groundwater modelling	^{90}Sr	Zuo et al (2009)
Dounreay	UK	Nuclear Power Plant	Soils developed on drifts from Caithness flagstones.	Coastal	Conceptual model. Limited discharges through deep GW due to engineered drainage	^{90}Sr, ^{137}Cs	Rostron et al., 2014

Sellafield		Nuclear Power & Spent Fuel Reprocessing	Sandstone Bedrock, overlain by drift deposits	Groundwater, Coastal & River	Geology & Hydrogeology. Groundwater well monitoring.	^{3}H, ^{99}Tc, ^{90}Sr, ^{14}C, ^{137}Cs	Holmes & Hall, 1980; Sellafield Ltd 2016; Chaplow, 1996; Black & Brightman, 1996;Michie, 1998; Littleboy, 1995; Bath et al., 1996; Heathcote et al., 1996; McMillian et al., 2000;
					Chemical associations of artifical radionuclides in cumbrian soils	Pu, ^{137}Cs, ^{90}Sr,^{106}Ru, ^{3}H, $^{95}Ar/Nb$	Livens & Baxter (1988); Peirson,1988
					Review of discharges & events to the environment from the Sellafield Site 1952-1982	^{137}Cs,^{241}Pu,^{90}Sr,^{106}Ru,$^{239/240}Pu$, ^{238}Pu,^{241}Am,^{3}H,^{99}Tc	Gray et al., 1995; Webb et al., 2006
LLWR - Drigg		Low level radioactive waste disposal site	Sandstone Bedrock, overlain by drift deposits	Coastal	groundwater borehole sampling and colloid soil analysis	^{3}H, U	Williams & Higgo, 1994; Warwick et al., 2002; Kwong et al., 2009
Hanford Site 300 Area, Washington State	USA	Nuclear Test Site	Alluvial Sediments overlying basalt bedrock	River	Hyporheic interactions of contaminated GW with the columbia river	U, ^{3}H, ^{90}Sr, ^{99}TC,	Gephert, 2010; Fritz et al., 2007; Johnson, 2016.
Savannah River, South Carolina		Nuclear Power Plant	Clays and alluvial sediments	River	Surface water samples collected in river basin from migrating groundwater contaminants from wastewater disposal in seepage basins	^{3}H, $1^{29}I$, ^{99}Tc	Beals & Hayes et al., 1995; Wam et al., 2012.
Barnwell, South Carolina		Low-level radioactive waste disposal site	Barnwell sands	buried trenches near to river basin and strip mine lake	soil, groundwater and trenches core samples of tritium to determine migration from land burial of low-level waste at Barnwell	^{3}H, $9^{0}Sr$, Pu	Czyscinski & Weiss, 1980; Zehner, 1983

Maxey Flats, Fleming Country, Kentucky		Low level radioactive waste disposal site	Weathered Shale & sandstone bedrock	47 burial trenches in flat topped ridges	hydrogeological, groundwater flow and radionuclide migration from trenches, tritium primary contaminant of concern	^{3}H	Zehner, 1983; Czyscinkski & Weiss, 1980
Coral Islands		Nuclear Test Site	Marine soils, consisting mainly of calcium carbonate, magnesium carbon, OC, N and P	Atolls	28 years study on radiological dose assessments including impact on groundwater	^{137}Cs, ^{90}Sr	Robison et al., 2003
Nevada Test Site		Nuclear Test Site	tuffs, rhyolites, tuffaceous alluvium	Desert & mountainous	Field scale migration of radionuclides	^{3}H, ^{14}C, ^{36}Cl, ^{99}Tc, ^{129}I	Tompson et al., 2002; Hu et al.,2008;
Amargosa Desert, Beatty, Nevada		Low-level radioactive waste disposal site	tuffs, rhyolites, tuffaceous alluvium	Desert & mountainous near River	Pore-water samples from test holes	^{3}H, ^{18}O, ^{14}C	Prudic et al., 1997; Striegl et al., 1996
Little Bayou Creek, Kentucky		PGDP plant (enriches U for use in nuclear reactors)	Carbonate bedrock overlain by fluvial-deltaic sediments	River, coastal plain	GW discharge along a channelised coastal plain stream/ springs	U, VOC, TCE,^{99}Tc	LaSage et al (2008a, b)
Sheffield, Illinois		Low-level radioactive waste disposal site		buried trenches near to river basin and strip mine lake	hydrogeology, groundwater flow and tritium movement along buried channel like depression	^{3}H	Foster et al., 1984; Garklavs & Healy,1986
Chernobyl	Ukraine	Nuclear Power Plant	eolian sands & some alluvial sediment at plume reaches	sandy aquifer	radionuclide transport in underyling sandy aquifer from burial trench & tracer tests (Cl-36)	^{90}Sr, ^{36}Cl	Dewiere et al., (2004)
				Dnieper-Bug estuary and the Black Sea	radionuclide fluxes into the black sea - saline mixing. Modelling: 3D time dependent flow and transport code	^{137}Cs, ^{90}Sr	Margvelashvily et al., (1998)

Lucas Heights Facility	Australia	Low level radioactive watse disposal site	Triassic. Hawkesbury sandstone overlain by shale, thin sandstone and siltstone lenses	River/Water divide between Mill and Barden Creeks	Evolution of mobile contaminant migration and conceptual model from buried waste in trenches	^{3}H	Hughes et al (2010)
Rhone Valley	France	5x Nuclear Power reactors	fluival deposits	Rhone River Delta	Direct discharges of radionuclides into river delta & coastal lagoons	^{3}H, ^{14}C, ^{134}Cs ^{137}Cs, ^{90}Sr, $^{238/239}Pu$	Jean-Baptise et al., 2018
Loire Valley		Nuclear Power Plant	fluvial deposits	Loire River	Predicting tritium migrating plumes	3H	Goutal et al (2008)
Unknown	China	Underground Nuclear Test Site	unknown	Aquifer - water supplies	Migration of ^{3}H as a tracer following underground explosions	^{90}Sr,^{137}Cs U, Pu, ^{3}H	Li-Xing et al., (1995)
Cheliabinsk Region	Urals	MAYAK plutonium production plants	River-bed sediments	Techa River system	Discharges to riverbed sediments of radionuclides. Anoxic conditions and behaviour of ^{99}Tc. Field sediment coring & lab experiments	^{99}Tc, ^{90}Sr, ^{137}Cs	Aarkrog et al;(1996)
Rhine River Basin	Germany	Nuclear Power & Reprocessing	River-bed sediments	Rhine River Basin	Discharges of waste-water	^{3}H	Mundschenk & Krause (1990)
Glatt Valley	Switzerland	Isotope Processing Plant	Glaciofluvial & quaternary aquifer of gravel and sand	River Basin	^{3}H as field tracer for surface & groundwater movement	^{3}H	Santschi et al., 1987
Paks	Hungary	Nuclear Power Plant	flat fluvial deposits	River Danube	experimental and modelling of tritium washout by precipitation	^{3}H	Kollo et al., 2011
Doel & Tihange	Belgium	Nuclear Power Plant	clay, silt and sands	River Basin	environmental risks from radioactive discharges	^{3}H, fission products	Vandenhove et al., 2013

Dessel		Low level radioactive waste disposal site	Boom clay, silt and sands	Wetlands & River Basin	environmental risks caused by contaminated groundwater (ERICA) and hydrogeological characterisation	^{3}H, fission products	Vives I Batlle et al., 2016; Beerten et al., 2010
Ignalina	Lithuania	Nuclear power plants/nuclear objects	Sandy Loam & Clayley loam	Lake Druksiai	regional tritium measurements in groundwater in four locations	^{3}H	Cidzikiene et al., 2014

2.4 Tabulated Summary of Archive Data

Over one hundred archived Sellafield Ltd documents are used to describe and reconstruct the history presented and data was extracted to build a long-term temporal picture of tritium occurrence at the Sellafield beach foreshore. The results are presented on the assumption that all tritium analysis was carried out in an accredited laboratory at the time by liquid scintillation method, although the exact methods and accuracy are not presented in many of the scientific reports. This is probably due to them being produced as an internal company records and methods were probably part of their management standards at the time (but copies were not found during the search). Although there is a range of documents spanning over forty years (1975-2010), when pieced together they form a compelling picture of the evolution of a tritium plume at the Sellafield beach. A summary of events and data have been reconstructed, re-interpreted and described in chronological order in Table 2.3 and the following sections:

Table 2-3 Summary of field investigations related to the beach springs area, near Sellafield site (1975-2010).

Date		Field Investigations/Events	Sources
1967	**i.**	Tritium readings in hand drawn graph for the Irish sea include readings for Seascale, Drigg and Braystones near the site, with peak concentrations at Braystone of 30 $pCiml^{-1}$	Unknown, 1967
1974	ii.	Tritium (^3H) recorded in the river Ehen between April to June with a range of 3.1 – 100 nCi/litre	Unknown, 1974
1975	i.	Routine sampling of sea water from the foreshore found elevated levels of tritium, along with an abnormal result in the mouth of the river Ehen this triggered further investigation of the beach and pipeline area to locate the source	Hetherington, 1975; Whittaker, 1977; BNFL Event record No002, 1975
	ii.	Natural springs/seepages found on the shore below high-water mark, approximately 100 metres from sea pipeline from the nearby nuclear site in December 1975	
	iii.	^3H activity for the freshwater component was recorded in the spring waters in the range of 0.6-0.8 microcuries/litre, with peaks of 1.0-1.5 microcuries/litre	
	iv.	<2 nCi/l very little tritium detected in the local sea water	
	v.	Extensive radiation surveys were carried out over the foreshore and the sand dunes near the springs & pipeline and no abnormal levels found	
	vi.	Pipeline was inspected and excavated up to 20 feet of overlying sand. No contamination or leaks were found	

1976	i. Possible sources of tritium at the Sellafield site (formerly called Windscales) were reviewed and identified several potential waste source terms. Major plant leaks were ruled out ii. Monthly tritium levels at the main peak discharge zone spring location were recorded iii. No ^{3}H was detected in the River Calder water samples iv. Daily & weekly samples taken seaburn sewers, effluent drains, old ROF drains and factory sewer no evidence to link them to the tritium found at the springs v. Detailed survey of the springs was made in October 1976 and verified highest samples from a narrow band near the pipeline vi. Additional boreholes were commissioned near potential source terms vii. V notch weir measurements of the spring flows were attempted but not successful viii. Institute of Geological Sciences (IGS) were engaged in October 1976 to investigate hydrogeological conditions ix. Early indications that tritium may be present in the riverbed of the Ehen.	Moore, 1976; Howells, 1976a;1976b; Longley, 1976
1977	i. Six holes were dug by JCB at regular intervals along the beach shingle front 170 metres south of the pipeline. Groundwater was found at a depth of 1.0-1.5 metres ii. Indication that some ^{3}H may occur slightly north of the pipeline, but most is centred about 140 metres south of the pipeline iii. Field observations over three months indicate large changes in beach topography, and a prominent spring/peak discharge zone is always seen draining this area iv. An experiment is conducted to monitor ^{3}H samples at the spring peak discharge zone at 30-minute intervals from high water until return of the tide 7 hours later. v. Estimates to ^{3}H flow rates from the spring peak discharge zone channel by constant rate injection method (violet dye tracer) of 0.8 curies/day vi. Flow rate measurements taken in the River Ehen and Calder on the area near the site by sudden injection method (integration of lithium chloride) during low tide, to avoid tidal effects. ^{3}H content measured throughout the day by liquid scintillation counting. ^{3}H in the range of 0.6-0.8 curies/day in the river Ehen and 0.03 curies/day in the river Calder. vii. Tritium arriving at the beach could be from several sources including waste disposal trenches and mortuaries and from more minor effluent and silo plant leaks viii. Samples of water from riverbed (river Ehen) showed high concentrations of tritium up to 3,300 nCi/l in area about 140-150m from the pipeline ix. Eight boreholes drilled into spit between River Ehen and the beach (BHP2,3,6,23,32,45,47) and monthly ^{3}H concentrations collected x. Variations in water level in the boreholes on the beach studied by IGS team using munro recorders xi. Water table maps of the beach area prepared by IGS xii. Large scale borehole drilling programme in flight across the site (60+ boreholes planned)	Whittaker, 1977; Paske, 1977; Greary, 1977; Luxmoore, 19779(a-h); Warren 1977(a-b)

1978	i. Distribution of ^3H in river Ehen riverbed mapped out from a 10m grid of sampling points ii. Pipelines under beach spit excavated and physically examined – no leaks found iii. Groundwater sampled alongside pipeline below high-water mark – no signs of unusual tritium, concluded no leak from pipeline iv. Leak tests performed on sea tanks (potential tritium source) – no significant results	Whittaker, 1978; Warren, 1978; Jones, 1978(a-c)
1979	i. Hydrogeological survey completes, and results published in IGS report 80/2 ii. Total of 55 boreholes drilled across site to understand groundwater chemistry iii. Six areas on site found with higher concentrations of tritium iv. Buried channel feature is described following borehole log analysis and possibly connectivity with the coastal area v. River and borehole water levels appear to respond to tidal and rainfall influences near the beach area vi. Interactions between the groundwater, river Ehen and the beach are described	Holmes & Hall, 1980; Holmes, 1979; Hall, 1979; Jones, 1979(a-d)
1980	i. Environmental impact of radioactivity in groundwater assessment concludes impacts were minimal since the main flow was from the works towards the sea. Radiation dose less than ICRP levels for continuous supply for drinking purposes ii. ^3H flow rates taken in November 1980, 0.16 & 0.15 Ci/day at 3 & 6 hours after high tide. iii. Samples of interstitial water from riverbed (Ehen) at 900 nCi/l were observed 140m south of the pipebridge	Hermiston,1980; Jones, 1980(a-b); Marples, 1980(a-f)
1981	i. Groundwater flow model developed (SWIP2) – groundwater flow from the site beneath the r. Ehen to the sand spit ii. Initial attempt to estimate Calder river flow rates by florescent dye dilution gauging technique to improve model inputs iii. Regular quarterly sampling of beach boreholes and main beach peak discharge zone spring	Marples, 1981(a-c); Bibby & Clifford, 1981; Smith & Huntley, 1982(a); Marples & Smith, 1981.
1982	I. Beach boreholes BHP23 ^3H activity increased from 23 nCi/l to 46 nCi/l and BHP47 from 198 nCi/l to 306 nCi/l II. Acid Recovery Plant discovered a leak to ground and likely a significant contributor to groundwater contamination at the beach. It was thought that a narrow plume was tracking south west along the gravel filled buried channel, adjacent to the pipeline. Boreholes close to this plant (BHP84,85,103) showed abnormally high ^3H to support this finding III. BHP85 closet to the Acid Recovery Plant tritium activity levels dropped to below detection level after plant repairs IV. ^3H concentrations in beach springs are in the range of 40 nCi/l to 1400 nCi/l. V. Beach borehole (BHP3) closet to the springs 3H activity declined from 2200 nCi/l in 1978 to 1000 nCi/l in 1982 supporting the two-plume theory and could be interpreted as indicating the trailing edge of the first plume VI. Further investigation of the sea tanks area is recommended as well as a potential leak near the uranium drum store	Oakes, 1982(a-d); Oakes 1983 (a-b); Maher,1982; Smith & Huntley 1982(a-b); Smith, 1982; Donoghue & Coverdale, 1982; Soil Mechanics, 1983; Jones, 1982

	VII. VIII.	^3H found under mortuaries in north group during environmental excavation (1.5-2.0 Ci) Geological mapping fieldwork of rock exposures in the River Calder, River Ehen/coastal area and on the site boundaries	
1983	I.	Quarterly ^3H monitoring of boreholes & beach springs	Oakes, 1983a-g
1984	II. III. IV.	Summary of current knowledge for river Ehen/beach discharge area is produced and two papers with recommendations for further programme of work to refine model parameters and understanding in this area. It is unclear whether any of this work was then carried out as no further reports were found Quarterly ^3H monitoring of boreholes & beach springs ranging from 100 nCi/l to 11 nCi/l on a seasonal basis, highest concentrations were found in the driest summer months, and rainfall dilution in winter months lowers concentrations. Flow logging in sandstone north of the site suggest that groundwater flow is dominantly through fissures, but a considerable distance from the beach area (>2km)	Williams, 1984; Oakes, 1984 a-d; Oakes & Coverdale, 1984; Robertson & Perkins, 1984
1985	i. ii. iii.	Quarterly ^3H monitoring of boreholes & beach springs Proposals written to investigate the river Ehen and beach discharge area – unclear whether the work was completed A review of the groundwater monitoring on the site concluded that the Windscale trenches were the main source of going 3H at the beach	Oakes, 1985a-d; Moore, 1985; Oakes & Jones, 1985; Moore, 1986a-b
1986	i.	Quarterly ^3H monitoring of boreholes & beach springs	Moore, 1987a-d
1987	i. ii. iii.	Quarterly ^3H monitoring of boreholes & beach springs BHP103 downstream of the main source area extended from 40.3m to 65.2 m to determine depth of tritium in the buried channel. Two zones of contamination were found in BHP103 with peaks at 0.24 mAOD and -19.36 mAOD	Davis, 1987;
1988	i. ii.	Quarterly 3H monitoring of boreholes & beach springs Geophysical surveys (seismic refraction & reflection) were carried out across the axis of the buried channel, in proximity of the seaward site boundary	Moore, 1988
1989	i.	Quarterly 3H monitoring of boreholes & beach springs	Moore,1990; Coverdale, 1990a-b
1990	i.	A review of the groundwater monitoring and investigations in the area of the disposal trenches	Poulton & Clegg, 1990; Moore, 1990
1993 - 1994	i.	Airborne thermal infrared survey of beach springs in the Sellafield area and field verification surveys. One spring north of the site recorded abnormal tritium levels towards the braystones station.	Hydrotechnica,1993
2003-2004	i. ii.	Geochemistry and Conceptual model development at the site, no specific beach spring/river Ehen studies Preliminary water balance for the Sellafield site	Sears & Thomson, 2003; Smith & Cooper, 2004; Hunter, 2004; Randall et al., 2004

2007	i. LIDAR survey as part of Cumbria coastal studies found there was a high degree of spatial and temporal change in beach and gravel bars from 2002-2006	Halcrow Group Ltd., 2007
2009	i. Geotechnical site investigation at riverbanks on Ehen for bridge construction ii. Review of geological faults at Sellafield	Garrison, 2009; Cooper, 2009.
2010-2013	i. Large borehole drilling project and update of the Sellafield conceptual and groundwater model (connectflow). No specific work carried out in the river Ehen/beach springs area ii. Updated to the Sellafield water balance 2012.	Garrick et al.,2010; Gordon, 2013; Serco, 2010

2.5 Evaluation of Archive Data Timeline

2.5.1 Discovery of tritiated water on the Sellafield Beach (1975)

The earliest record found related to tritium in the Sellafield beach area is a hand-drawn graph with readings in the Irish sea during October 1967 by an unknown author. Peak readings were recorded at 30 $pCiml^{-1}$ near Braystones, which is equivalent to approximately 1000 Bql^{-1}, for comparison this is significantly higher than modern background levels in seawaters close to the site of around 20-40 Bql^{-1} in 2010. Tritium was first discovered at the Sellafield beach in December of 1975, following some routine environmental water sampling from the sea on the foreshore at Sellafield during 1975 and found elevated tritium levels compared with other samples from the sea near the site (usually at 0.3-1.0 $nCil^{-1}$ near the pipeline) , they also had found one sample with an abnormal tritium level in the mouth of the River Ehen in 1974 that lead to an investigation to find the source of this contamination (Hetherington, 1975). An BNFL operational event report was raised and recorded as Event No0021 and reported to the nuclear regulators. This was a reportable International Nuclear Event Scale Level 1 (INES1)-anomaly beyond the authorised operating regime (Webb et al., 2006). The Event report describes a '*number of issues of water on to the beach at Sellafield containing tritium, at a concentration 1 micro Ci/litre were detected'.* This very early report does not describe how far north and south along the beach that samples were taken but does provide an indication of the spatial footprint describing the area of upwelling to be an area of 70m by 70m at an estimated distance of 100 metres south of the pipeline and would accord with the current hot spot discharge found in present day environmental monitoring data (shown in Figure 2-5).

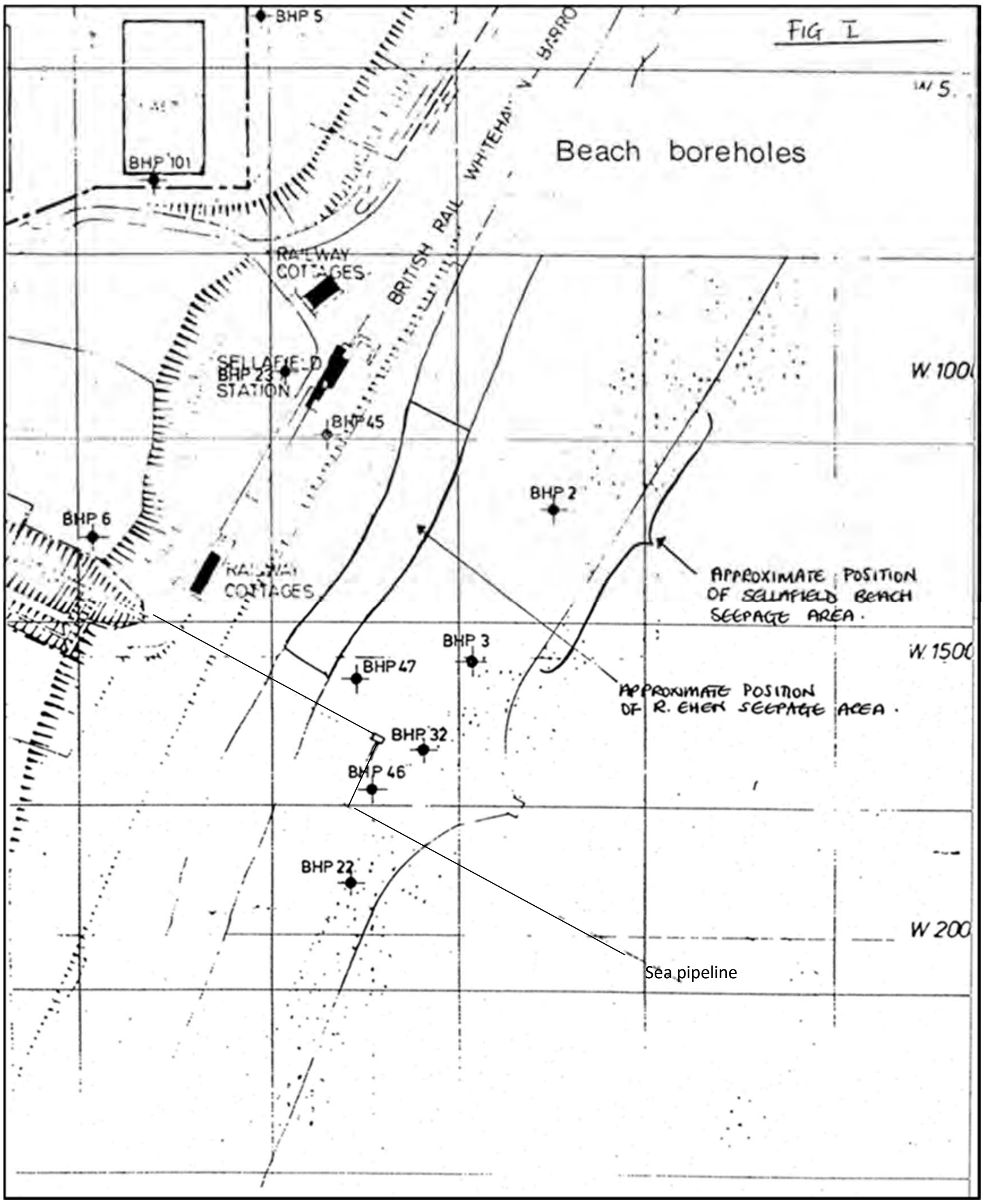


Figure 2-5 Early map of the beach area where the seepage of tritium was found taken from (Oakes 1998).

In late December 1975, an inspection of the foreshore at Sellafield found several seepages of water on the shore (referred to as the 'confluence') below the high-water mark and samples taken from pools on the beach at Sellafield showed tritium activity levels of about 100 $nCil^{-1}$. When corrected for salinity, recognising the brackish samples were a mixture of fresh groundwater and high salinity sea water, the tritium content of freshwater was in the range of 800 to 1600 nCil^{-1}. The peak discharge zone spring (hot spot) upwellings gave consistently higher levels than others and since first detected the specific activity is generally in the range 0.6 to 0.8 µCil^{-1} peaks of 1.0 to 1.5. Initial thoughts at the time were that the source of the tritium was associated with a direct leak from the effluent sea pipelines cutting across the beach about 100 m north of the spring tritium hotspot discharge. The possibility that the source was discharge of contaminated groundwater moving under the site only more gradually gained traction over 1976 was not particularly foreseen, recalling in that era migration of contaminants in groundwater had hitherto received little research attention globally and was an emergent subject area. There was very little tritium detected in the seawater close to the Sellafield beach (< 2 nCil^{-1}) so therefore tritium from direct sea pipeline discharges being washed up onshore was discounted as the cause of the beach discharges.

In December 1975 extensive radiation surveys over the foreshore and sand dunes in the vicinity of the percolations and the pipelines showed no abnormal levels. There was no evidence of associated fission products. An inspection of the pipelines was also undertaken. This entailed excavation of the overlying sand to a depth of up to 20 feet and no leaks were found. Samples of sand taken from below the pipeline were not contaminated and radiation were normal. The mechanical cladding of the pipeline was in good condition.

2.5.2 Early investigations of tritiated waters in the beach area (1976-1980)

A number of reports cover a range of field investigations related to the discovery of tritium in beach springs that were carried out between 1976 and 1980, studies took place in six main areas close to the Sellafield site: (1) the Sellafield Beach, (2) Boreholes on the nuclear site (3) the River Ehen, (4) Spit of land between the River Ehen and the Beach (5) the east bank of the River Ehen and (6) the old effluent sealines.

Monthly sampling of the water percolating on the beach started after the discovery of the tritium on the beach in 1975. Howells (1976) presents monthly tritium levels throughout 1976 and indicates that all samples of water taken from a similar location on the beach show abnormal levels of tritium, although no coordinates or exact maps are provided. It is difficult to also understand what 'abnormal' levels of tritium are, there is no quantified threshold level quoted. The salinity was reported in the range of 40-80% showing the presence of some freshwater. When the tritium levels in the samples were corrected to freshwater component, they were in the range of 200 to 1,500 $nCil^{-1}$. In parallel over the year of 1976 monthly samples were collected across the site in a number of the drains to check there was no obvious signs of leakage from the plants, and it was concluded that there was no evidence that the source of tritiated waters was from the drainage system, in most cases the levels had been low. The tritium levels in the sea were around the expected levels due to permitted effluent discharges via the sea pipelines (0.3 – 1.0 $nCil^{-1}$). The factory sewer (a potential direct pathway to the beach) was checked by the government agency (MAFF) and no regular discharge of tritium was found. Water samples taken from the River Calder did not detect any tritium. Only one of the Windscale boreholes (at the northern part of the site) at the time showed the presence of tritium.

The area around the sea pipelines were excavated along the pipeline towards the site and a physical examination carried out that revealed no evidence of any broken pipe or effluent leakage. Investigations so far had established that this tritiated beach water did not originate from a leakage out of the sea pipeline. The cause was now suggested to be contamination of groundwater on the

Windscale site. At this stage they did not know whether the origin was fission products including tritium or just tritiated water free of other contaminants.

A detailed survey of the 'springs' of water on the beach south of the pipeline was made in October 1976. This verified that the highest concentrations of tritium are in samples taken from a narrow band across the beach. The levels further south rapidly diminished (Howells, 1977). Crude estimate of the beach springs discharge suggests a flow rate of 10 litres per minute or about 14-15 m^3d^{-1}, which is estimated to be around 14-15 $mCil^{-1}$ or just under 0.5 Ci/month. Rough measurements of the rate of tritium seepage into the River Ehen were carried out on the 5th April 1976, shortly before the equinoxial spring tide of 9.9m. Samples were taken 300 metres downstream of the pipe-bridge and 200 m upstream (for a background measurement). The experiment recorded a high tritium seepage rate of an estimated 0.84 Cid^{-1}. The rates of tritium seepage onto the Sellafield beach and into the River Ehen were found to be approximately 0.8 Cid^{-1} each, making a total of 1.6 Cid^{-1}, or about 860 curies over the one and half years that observations took place. The seepage is presumed to be coming from the bed of the river roughly 150 metres downstream of the pipe-bridge. Consequently, the investigators hypothesised that there was a distinct possibility that the seepage rate would be controlled (and even reversed) by the hydrostatic pressure on the bed of the river as a result of varying river flow (Howell, 1977). Since this part of the river is also tidal, high spring tides could have a similar effect.

A review of site operations (leakages) did not reveal any likely source (Moore, 1976). Health physics personnel tried to assess the movement of tritium into the ground, and the report is a little confused around expecting to find fission products at the beach, however references to other similar nuclear sites with groundwater plumes in the USA suggested that fission products would be held up in soils close to buildings and would not have moved far in the groundwater. The Institute of Geological

Sciences (IGS) was then engaged by the site operators to provide specialist hydrogeological support. It was thought that the tritium activity passed under the River Ehen before surfacing under a hydrostatic head onto Sellafield Beach. The action following this report was to sink additional boreholes on the seaward side of the Windscale site to attempt to pinpoint the possible source activity.

An interesting fact in this report is that there was no reduction in the tritium concentration in the boreholes on the site during a period of an industrial dispute which lasted from the 28th January to the 13th March 1976, during this time the main reprocessing plants were shutdown, with known effluent streams containing tritium significantly reduced. There was no obvious increase in the tritium concentration was observed when the plant was restarted either, which may have indicated that there were no ongoing leaks from the main process plants. The shutdown period was only three months however and if it had been a slow leak over a prolonged time, the tritium levels may not have necessarily responded immediately.

Whittaker (1977) and Paske (1977) report on an investigation to take preliminary measurements of the rate of tritium seepage onto the beach and into the River Ehen & Calder. A comprehensive assessment of the leakage of fission product activity from the one of the main silos and a rough initial assessment of the quantities of waste tritium arising from the 1955-1962 tritium production was also reported.

On January 17th, 1977 a mechanical excavator was used to dig six holes at regular intervals along the beach to 170 metres south of the pipeline. The holes were dug at the base of the steeply shelving shingle in a straight line parallel to the shore (Figure 2-6). Groundwater was found at a depth of about 1.0 to 1.5 metres and samples were taken. Groundwater samples in the dug holes reported 93-99%

seawater. This was different from the surface water drawing from the cobble area and suggests that seawater may be concentrated in the shingle zone by evaporation between tides. No attempt was made to relate the tritium concentration in the groundwater to the freshwater due to this potential effect. Tritium concentrations ranged from 5.0 to 180 $nCil^{-1}$ in samples found along the beach. Whittaker (1977) indicates that the tritium-bearing aquifer is centred about 140 metres south of the pipeline and is comparatively narrow, the width at half maxima being only 40 metres. A slight rise in activity near the pipeline is suggestive that a "second aquifer" exists on the north side of the pipeline, confirming results obtained earlier by Longley (1976), who found tritium levels of 20 $nCil^{-1}$ at 75% salinity in a spring 100 yards north of the pipeline. It could also be interpreted as one aquifer with the effect of the heterogeneous deposits and narrow plume behaviour.

Observations made over a three-month period indicated that there can be big changes in the topography of the beach from day to day. However, normally there is only one spring draining the whole area from the pipeline to 200 metres south of the pipeline. Only rarely were there two or more springs. It was thought a reasonably accurate measurement of the total rate of tritium seepage in this 'southern' aquifer could be obtained by measuring simultaneously the tritium concentration and flow rate in that channel. Therefore, an experiment was carried out on the 17th January 1977, roughly midway between neap and spring tides to study the short-term variations in tritium concentration and salinity in the spring discharge area. The water was sampled from the spring discharge area at a point roughly 20 metres downstream of the peak discharge zone in order to ensure good mixing.

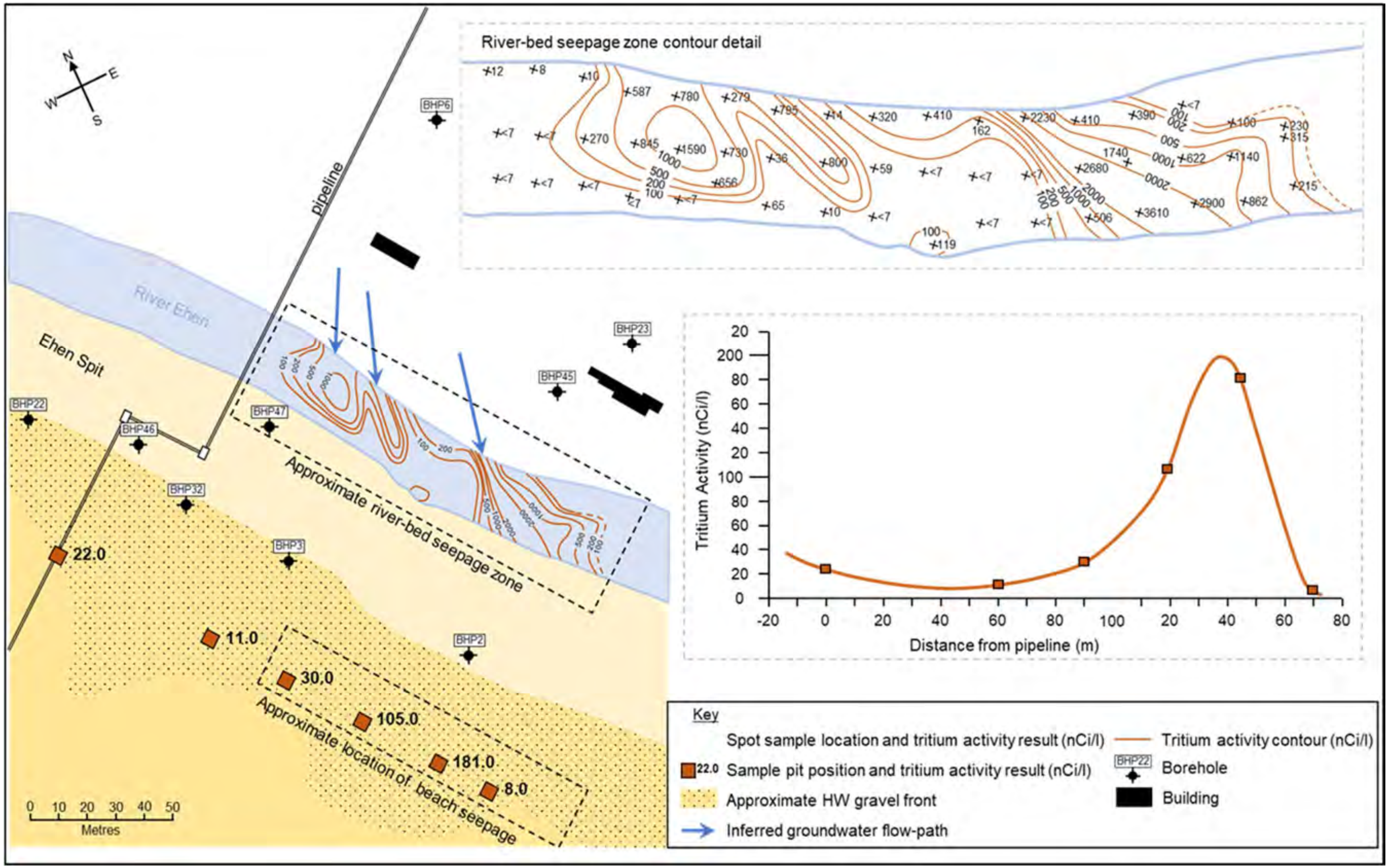


Figure 2-6 Spatial Distribution of tritium activity in river-bed and beach seepage waters, and locations of beach boreholes drilled in the late 1970s (BHP2;BHP3;BHP32;BHP46;BHP22;BHP47;BHP45;BHP23;BHP6). Reproduced and collated from (Whittaker,1977;1978; Smith, 1982 and Oakes, 1985a)

It was seen that the tritium activity in the brackish water rises from low level of less than 100 nCi/l shortly after high water, to an asymptote of 365 nCi/l about half an hour after low water (Figure 2-7). When the tritium activity is referred to the freshwater component, the concentration was initially constant at about 950 nCi/l falling to about 750 nCi/l by the time the tide returned. Gradual changes in tritium is probably due to varying dilution by rainwater run-off.

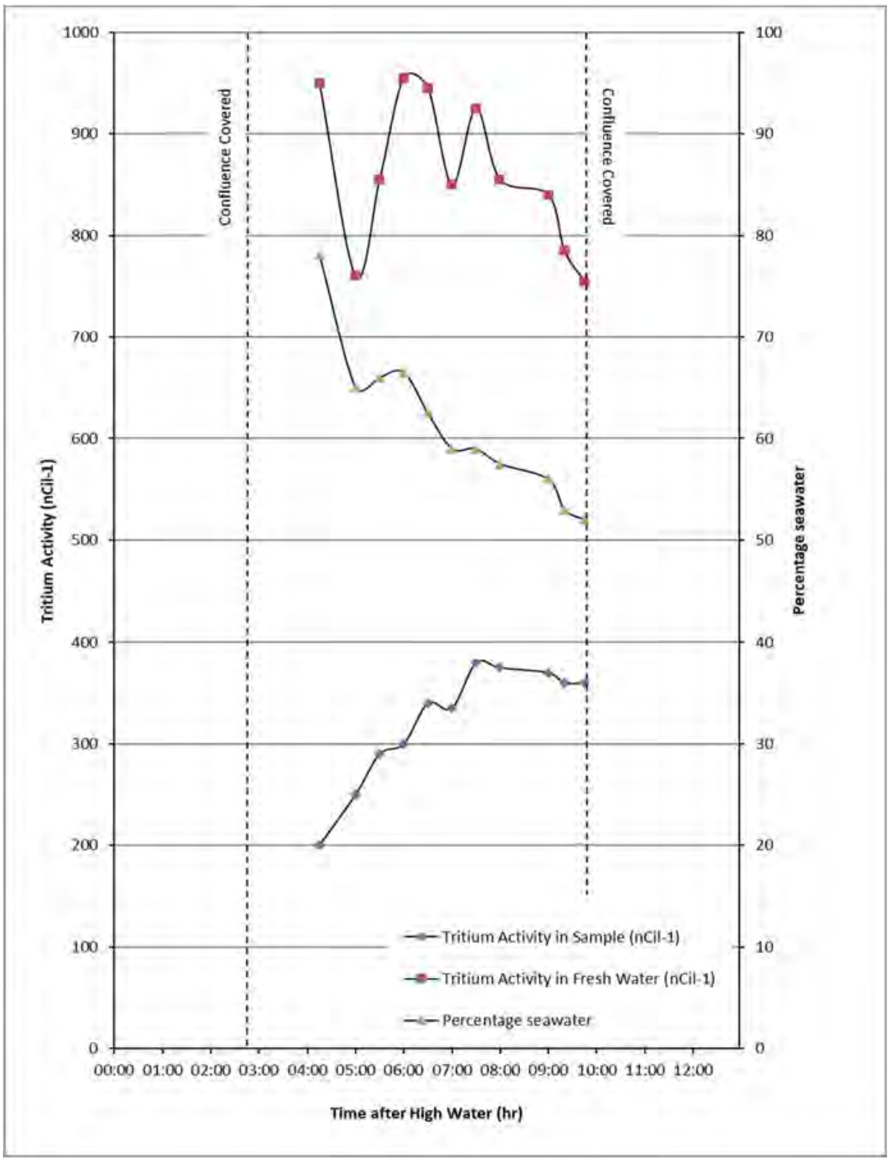


Figure 2-7 Reconstructed from Whittaker (1977) from the first experiment carried out in 1977 between the high and low tides. 'Confluence' refers to the beach spring discharge.

Early attempts to measure the springs flow rate did not work with a slotted weir due to the scouring effect of the high flow on the sand, resulting in undercutting of the weir and bypassing of the flow. A 'constant rate injection method' was selected instead using methyl violet dye as the tracer during low tide (Paske, 1977). The experiment indicated the tritium flow rate remains reasonably constant whilst the tide is out. However, on the return of the tide, the increasing hydrostatic pressure must inexorably cause the flow to cease and eventually reverse itself, especially during spring tides. If the flow was stopped and reversed and retained in the aquifer, the 'held-up' tritium would potentially be released at a higher rate immediately the tide had fallen. Tritium flow rate was estimated at 0.78 curies per day. It should be noted that this value is more than 50 times greater than the earlier estimate reported by Howells (1976). It is thought that this rate was measured near neap tides when the bulk of the water was flowing beneath the sand.

Whittaker (1977) linked the dilution of tritium with rainfall but also suggested that when the River Ehen is in flood, the greatly increased hydrostatic pressure on the bed of the river may cause water to seep into the underlying aquifer, thereby reducing the tritium concentration. Thus, variations in the level of the River Ehen could also be a contributor to the tritium fluctuations.

Following various onsite investigations of the plants and operational areas, it was confirmed only a few minor plant leaks were identified and the inventory assessments would not have been of enough magnitude to give the tritium activity readings at the beach. Notwithstanding the small contribution, the assessments of total loss to ground were made, and the main leak would have likely started in August 1972 following an extreme temperature excursion in one of the silo compartments. Now that a major process plant leak had been ruled out, a further assessment of potential tritium source terms at the operational site was carried out (Whittaker, 1978), and it was found that there was tritium waste stored in several areas on the site from around 1955 to 1962. The majority of the waste was tritium rich furnace linings that were disposed of either to (a) an on-site burial trench (b) the dry silo and (c) two mortuaries situated at the northern end of the site. These linings contained the bulk of the tritium waste from the early tritium production programme. There was also known accidental losses to the atmosphere in the period 1958 to 1962 due to a silo fire and leakage from some mortuaries. The bulk of the inventory was concluded as being contained in discarded furnace linings wrapped in polythene wrapping in the concrete mortuaries and buried in the Windscale trenches, and this was possibly the main source of tritium surfacing on the foreshore. The mortuaries on inspection were intact and a pathway to groundwater was ruled out.

Sixty groundwater monitoring boreholes were drilled between 1975-1977 to sample groundwater, six of which were drilled along the Sellafield beach and the River Ehen Sandspit (Figure 2-6) and monitored on a monthly basis (BHP22; BHP46; BHP32; BHP47; BHP03; BHP02). The data observes an anomalous high result of 2170 $nCil^{-1}$ in the groundwater monitoring borehole number 3 (BHP3) closest to the beach seepage area. Drive point samples of water from the riverbed of the Ehen were taken in June 1977 and showed high concentrations of tritium up to 3,300 nCl^{-1} in a broad area roughly 140-150 metres downstream of the pipe-bridge (opposite the railway station building). Further samples were taken from the riverbed in May 1978 (Figure 2-6) and mapped across a detailed 10m sample grid (Whittaker, 1978). The maximum concentration observed during this investigation was 3600 $nCil^{-1}$ at a point 140 metres downstream of the pipe-bridge on the west bank of the river back in 1978 were probably real and the riverbed and beach contamination are ‘in line’ with each other on a south-westerly trajectory from the main site area. High nitrate concentrations were observed in the boreholes nearest the pipelines, possibly indicating links with nitrate rich effluents from plants.

Three boreholes (BHP06; BHP23; BHP45) were also drilled on the east side of the riverbank on Sellafield site side of the river (Figure 2-6) but only had low mean tritium levels in comparison to the riverbed and the sand spit with results between 3.0 – 40 $nCil^{-1}$. The sea pipelines were carefully inspected and there was no evidence that a leak was found, and soil samples taken from underneath the pipeline were clear of activity. The mean tritium concentration in the south pipeline (averaged over 3 years) is 43,500 $nCil^{-1}$ and this would be high enough to explain high concentrations in the Ehen river bed however the leak rate would need to be 32 m^3d^{-1} (700 gallons) to produce the levels found in the river bed samples. Since pumping is restricted to a few hours per day the leakage rate would need to be higher and a leak of that magnitude would be visually noticeable, and no such areas of leakage were found along the pipeline. It was concluded that the pipelines were not the source of the tritium on the beach.

The observed seepages of tritium in the Ehen riverbed and onto Sellafield beach appear aligned and could potentially suggest a similar source origin and the plume accounts for both, the failure to detect the plume in the groundwater monitoring well on the east bank of the river (the site side) suggests a deeper pathway with a rise to the beach receptor discharge points flowing under the west bankside monitoring well (beach side). However, the plume discharge points are quite discrete and if a shallow plume is narrow the near bankside monitoring locations may have missed it. Figure 2-6 includes inferred groundwater flow path to the river interpreted by Whittaker (1978) at the time, the pattern of tritium distribution in the river could also be controlled by the permeability of the riverbed as the pathway rises to discharge points. There is also a 'buried channel' feature mentioned in the reports and this may also be a contributing feature where channelling of higher flows along a discrete pathway may look possible.

The concentration of tritium when expressed as a freshwater component was found to vary on the beach and was well correlated to rainfall, being lowest after a period of heavy rainfall. Monthly mean tritium concentrations show a minimum of about 250 $nCil^{-1}$ in the winter and a maximum of about 1200 $nCil^{-1}$ in the summer. Flow rate measurements were carried out in the river and on the beach springs in 1979. Flux rates were estimated at 0.6 Ci/day in the river and 0.8 Ci/day in the beach springs. This would indicate an arising of some 500 Ci per year.

The hydrogeological investigations concluded in 1979 and were published in Holmes & Hall (1980). It was concluded that values of hydraulic conductivity could vary over a very wide range over short lateral and vertical distances in the unconfined glacial deposits, but groundwater flow in sandstone bedrock was through fractures with lower more constant hydraulic conductivities and is more semi-confined in nature, and a complex flow horizon is observed at the boundary between the glacial deposits and the sandstone.

The hydrogeological relationship between the aquifers, the River Ehen and the beach (sandspit) are complex. The river runs parallel to both the coast and the water level contours of the aquifer. Local groundwater flow regime was interpreted to be dependent upon the tidal state of the river Ehen (Figure 2-8). Extra high tides can raise the water level of the river by over 2 metres at the pipe-bridge and at high tide, river water is recharged into the sandspit as bank storage. Groundwater also continues to discharge seaward beneath the river, in the glacial deposits. The River Ehen forms an aquifer boundary as groundwater from the site flowing into the river from the glacial aquifer are potentially separated from the spit groundwater for at least 10 hours of every tidal cycle. Howells (1979) describes differences in conductivity and nitrate values of groundwater between the sand spit and the site side of the river. This water contains significant levels of nitrate and tritium and indicates a link between them in the plume. Tritium also emerges in bed of the river in an irregular distribution which may indicate preferential pathways, alongside water level fluctuations in all boreholes in this part of the site in response to tides. It was concluded that the time lag exhibited in the water level responses to tides, the chemical differences between groundwater on the beach and the site, and the isolation of the beach groundwater from the site glacial aquifer by the river Ehen, all suggest that beach groundwater may be derived from the sandstone bedrock, or possibly from groundwater moving in the base of the glacial deposits (under the river).

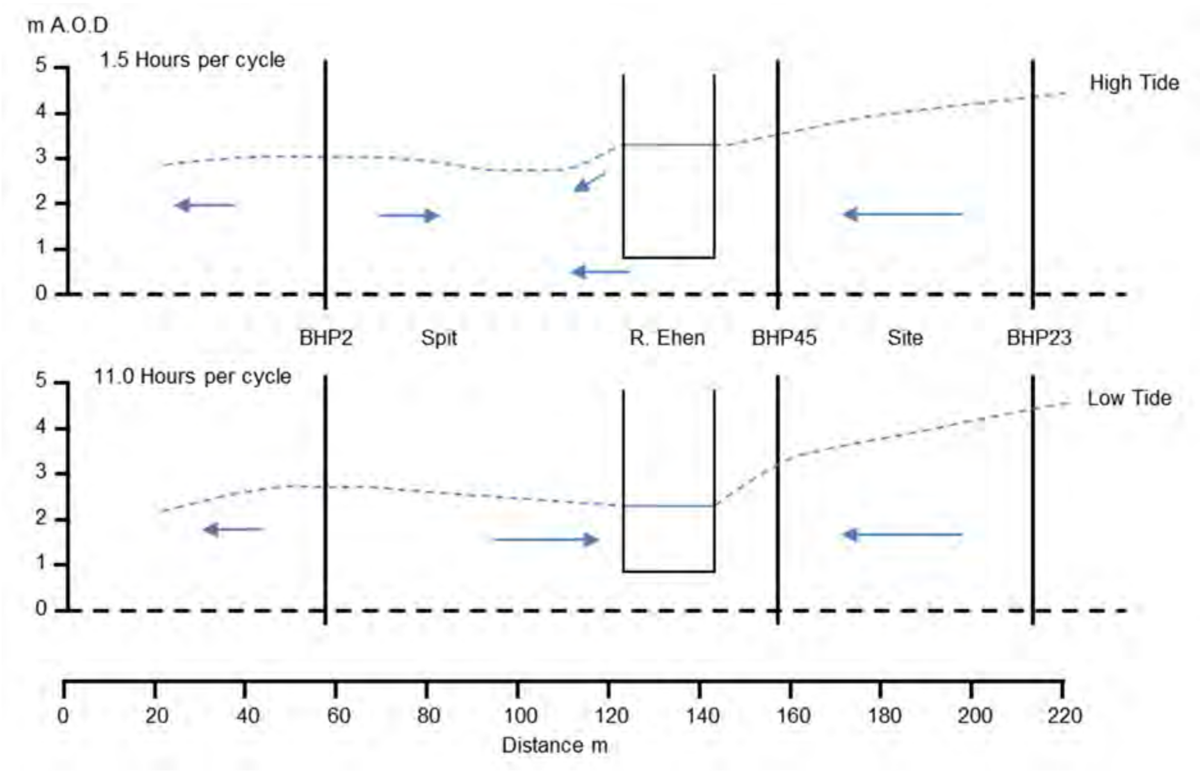


Figure 2-8 Interpretation of water level differences between Ehen Spit, River Ehen and the site glacial aquifer with an indication of possible flow directions reproduced from Holmes & Hall (1980).

Monitoring of the contamination expression in the River Ehen riverbed only received investigation in the early years of investigation (1977-1979) and its expression since can only be gauged by the concentration monitoring of the river itself. The early study points to a complex plume discharge with potentially the same plume able to discharge to both the river Ehen and the beach springs and reversal of flows (due to tides occurring) and a possible complex relationship of flow and contaminant exchange between the river and aquifer towards the beach, further investigation of this complexity (although recommended in the 1970s-80s) was not undertaken. Early measurements suggest the tritium discharge to the beach and the riverbed were similar and has not since been investigated and was based on limited late 1970s data.

2.5.3 Hydrogeological and source zone investigations related to the beach area (1980-1990)

Regular quarterly monitoring of the beach seepage, rivers, beach and site boreholes were carried out for most part of the 1980s. Regular environmental impact assessments were carried out based on the data collected, all of which concluded that radioactivity measured in groundwater at or beyond the Windscale site perimeter would produce radiation doses to the public well below the limits recommended by the International Commission on Radiological Protection (Jones, 1980). Two types of radionuclide transport were considered in these studies: migration in the unsaturated zone above the water table, and migration in the saturated zone. Techniques to further characterise the plume at the perimeter and beyond such as tritium/helium ratios for groundwater ageing or determination of long-lived highly mobile fission products such as technetium-99 and Iodine-129 could give indication of migration pathways and suggests further work (Marples, 1980b; Crowder, 1980), but records are unclear if this was ever completed and no reports on this suggested further work were found during the archive search.

Work commenced at the end of 1980 to develop models of groundwater flow and radionuclide transport at the Sellafield site. US geological survey code SWIP2 was selected and the geological and groundwater flow model was built and calibrated using the field data collected from 45 boreholes across the site (Bibby & Clifford, 1981). The final modelling output reports were not unearthed during this study, but later work on groundwater modelling of the saline interface and the Sellafield site was published in Heathcote et al., (1996).

A borehole west of the Acid Recovery Plant showed a tritium peak in 1979 and a gradual increase to 1981. A leak to ground was discovered in 1982, and likely started around 1972 according to the three leak events recorded in plant logs. The Acid Recovery Plant was concluded as a likely source of tritium in groundwater as the concentrations in the blending house were high enough to cause the arising if enough volume of liquor had leaked. The hydrogeological survey work indicated that the spilled tritium from this plant would flow towards the Sellafield beach. Plant repairs were carried out immediately in 1982 to stop the leak to ground (Maher, 1982). Smith & Huntley (1982b) suggest that the groundwater model predicted a high groundwater flow rate in the order of 4 metres a day and therefore a travel time of two years from the recovery plant to the beach, and now that the leak had been repaired they would expect to see a step change decline in tritium activity levels in boreholes near the plant within two years. These conclusions are slightly at odds with that reported by Oakes (1982b) suggesting that tritium from the acid recovery plant was already arriving at the beach.

Oakes, 1982(b) and Dongahue & Coverdale (1982) are key papers as they assess three potential contaminant transport scenarios to result in tritium contamination at the beach based on different source inputs. The main conclusion presents a two-plume theory to account for both the tritium levels on the beach and in the boreholes on site near the acid recovery plant and the legacy silo leak. The pathway is described by following a valley beneath the site in the St Bees sandstone which slopes towards the sea and is filled with high permeability sediments, with the axis of the valley coinciding approximately with the location of the pipeline. Further corroboration of this was that borehole BHP6 between the site perimeter and the river Ehen was absent of tritium and indicated that the first plume had passed prior to the borehole being installed. The groundwater velocity was calculated as 0.24 feet/day (0.07 m/d), which would need further calibration by field checking by pumping/packer testing of existing boreholes by the IGS. The modelling used a source term emplacement of 1958 (low level waste trenches) for 6 months to give the results seen at the beach in 1975. The two-plume model fit well but the report did note that the model was sensitive to the assumed source term activity. As the tritium levels were in decline from 1975 onwards at the beach, the plume would have arrived prior to its discovery in 1975. Given the high levels of tritium in some of the boreholes still on the site the second plume may have not reached the beach at the time of writing the report in 1982. Jones (1982) reported at a site survey meeting that 1.5-2.0 Ci of tritium had been found under the building of the tritium mortuaries at the northern end of site. If this was the case, there should be an increase in beach springs at some point in the future to indicate arrival of another plume. There was also a recommendation to sink more boreholes to help define the width of the plumes and indicates that the spatial characteristics of the plume were not fully understood at the time.

Smith (1982) mentions some spatial aspects of the beach springs as being 200-300 metres south of the pipeline and 250 metres west of the River Ehen. The geometry of the seepages is described as variable appearing as two distinct flows which merge as they flow seawards and on other days as a single broad discharge. The variations being in part due to the shifting topography of the beach under the influence of the sea and water. Soil Mechanics (1983) were hired in 1982 to identify and map all rock exposures within and adjacent to the site boundary. The study concludes that sandstone bedding dips at between 5° and 30° to the south-west and attains a thickness of approximately 800 metres at the coast. Faults were found in the St Bees Sandstone in the river Calder all striking WNW-ESE with consistent bedding. A deep infilled channel is described which formerly drained in a south-westerly direction and is cut into the surface of the sandstone. The channel appears to deepen from 20 metres at the north-eastern end of the site to over 45 metres at the coast. The river Ehen had limited rock exposures in the riverbanks, and there was considerable disturbance in the area as landscaping of material removed from elsewhere on site (Thorp construction project) onto the Ehen spit had just began.

Williams (1984) recognised that there is a distinct lack of information in the Ehen/beach discharge area surrounding the Sellafield site. The report summarises geological and hydrogeological information to date and recommends a further programme of work necessary to remedy these data deficiencies, although no further evidence that this work was completed was found. Interestingly it is suggested to carry out an intensive sediment and groundwater sampling survey along the riverbed in the river Ehen in order to detect whether radionuclides are being released at the surface however, doesn’t acknowledge the 10 metre grid drive point sampling carried out by Whittaker (1978). Further borehole drilling and geophysical surveys to define geology over the wider area, and hydraulic testing were also recommended.

Oakes (1985) also summarises the progress in investigations on the tritiated groundwater on the Sellafield beach over the previous ten years and proposes new hydrogeological fieldwork and concludes that effort is now being concentrated on identifying the specific tritium source terms on the site. At this point it appears that any further investigations into the beach discharge area are not completed, or the reports are missing from the archives. A review of the groundwater monitoring on the site (Oakes & Jones, 1985) attributed the ongoing tritium activity in the groundwater surfacing at the beach to be the Windscale trenches.

A key borehole (BHP103) directly down gradient of the Windscale trenches and centred in the buried channel was chosen to be extended from -19.0 mAOD to -45.0 mAoD (Davis, 1987). The channel was filled with coarse permeable sands and gravels and would likely provide a preferential migration pathway for groundwater and any contaminants to migrate towards the beach. Tritium depth samples were taken during drilling and found two main zones of peak tritium concentrations (Figure 2-9). The first in the glacial sand deposits at around 0.24 mAOD and the second at the base of the glacial deposits within the sandstone at approximately -19.0 mAOD and in direct hydraulic continuity. Below -26 mAOD in the sandstone all the tritium activity results were below analytical limit of detection. The depth profile supports the multiple plume theory given the near proximity of this borehole (BHP103) to the trenches, the second peak in the sandstone aquifer could indicate tritium source(s) migrating from further upstream of the disposal trenches. Identifying layers of contamination when drilling groundwater monitoring wells is difficult so even though this particular borehole profile appears to give two horizons, it should perhaps be treated with caution as smearing of contamination can happen whilst drilling and retrieving samples, this was the only borehole at the time in the area and others around the area during later drilling didn't produce such a prominent dual layer horizon for the tritium.

The characterisation of the deeper radionuclide groundwater contamination remains an issue at the Sellafield site, as it is easy for anomalous results to be produced at depth especially when drilling through shallow contamination in the drift deposits. A geophysical survey was also carried out in this general area of site. Both seismic refraction and reflection surveys were carried out across the axis of the buried channel, postulated from the borehole data and the three transverse lines extended towards the seaward site boundary (Moore, 1988). The traverse survey lines were close to the sea pipelines, so the seismic sources were restricted to cartridge gun devices, instead of the traditional small explosive to reduce interference. The refraction survey could not define the bedrock. The processed results showed the reflector surface (sandstone bedrock) at a depth between 30 and 40 mbgl which agrees with bedrock with borehole logs in the area and indicated a buried channel feature with axis running approximately east-west, parallel to sea discharge pipeline. The base of the channel was deeper than anticipated at -38 mAOD (approximately 58 metres below ground level) compared to -25 mAOD to -30 mAOD from borehole log interpretation.

Quarterly monitoring of tritium in boreholes and beach springs continued throughout this period, and the decreasing concentrations of tritium arisings on the Sellafield beach are consistent with a simple model of a large discrete disposal inventory followed by radioactive decay and by migration losses via groundwater. The magnitude of the tritium source in the trenches would mask any other smaller source term signatures at the beach discharges.

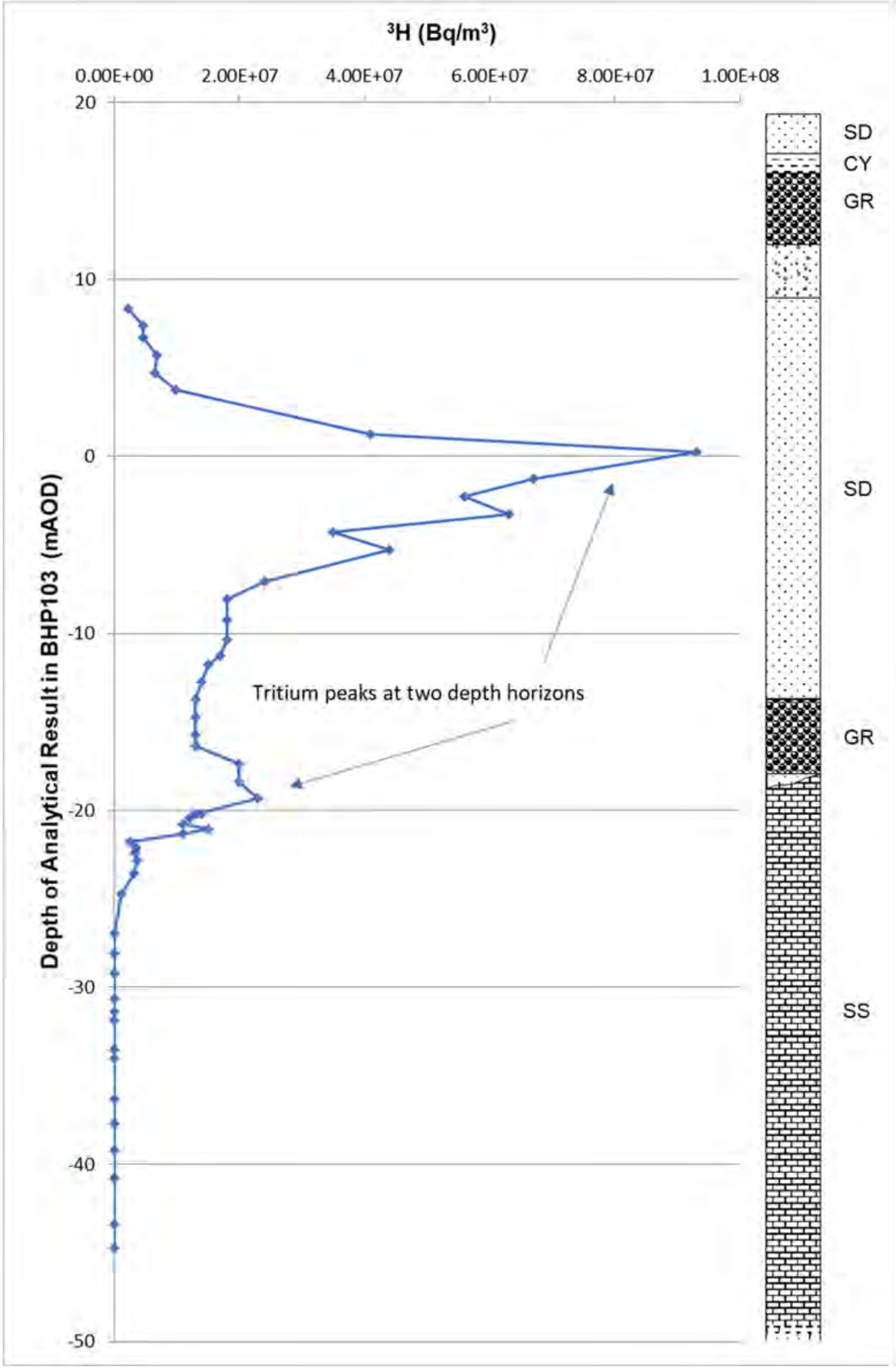


Figure 2-9 Depth profile of tritium results from BHP103 drilling log in 1987, located down gradient of the Windscale trenches two horizons of tritium peaks suggest more than one tritium plume is migrating in both the glacial drift and sandstone aquifer with direct hydraulic continuity, or possibly tritium migration caused by an artefact of the drilling process. (SS: sandstone; GR: gravel; SD: Sand; CY: Clays. reproduced from Davis, 1987)

2.5.4 Airborne thermal infrared survey of beach springs (1993 – 1994)

As part of a wider hydrogeological investigation during the NIREX project for deep radioactive waste repository geological investigations in the Sellafield area in the 1990s (Nirex, 1993), an airborne thermal infra-red survey was carried out in April 1993 and March 1994 of the intertidal beach zone to try and identify areas of groundwater discharges. A follow up field investigation to the identified areas was then carried out as part of the survey. Two phases of investigation are recorded in Gibbs (1995) and Hydrotechnica (1993). Phase 1 of the aerial thermal infrared survey, was carried out in a Piper Aztec aircraft using a daedalus DS1230 linescanner coupled to an SE labs 7000 tape recorded using 0.5-inch Aaipox tape. Flight lines were flown parallel to the coast and up to 2km inland to try to identify areas of onshore groundwater discharge, the survey was flown from Nethertown in the north to Drigg point in the south on the 11th March 1993. The areas identified from the survey were visited as a field verification the following month in April 1993. The sites were briefly assessed and surveyed, and samples were taken of the spring water discharges and analysed for a variety of determinands (major and minor ion concentrations, oxygen, hydrogen stable isotope ratios, tritium analyses and radiological measurements). The samples were taken at sites with low electrical conductivity readings (<47,000 µS/cm). A second field survey (Phase 2) was carried out the following year in March 1994 to determine the extent of movement of the beach springs by re-surveying the positions and to re-sample any discharges of interest. This survey also included a new survey area south of Sellafield site.

Considerable thought was put into the ideal conditions to gain maximum thermal contrast between the ground and surface waters. February/March spring discharges are usually at their maximum at this time and vegetation cover is minimised, in particular maximum thermal contrast occurs on clear cold days in the hours before dawn, so the ideal time was therefore a combination of pre-dawn coupled with a low tide exposing the maximum amount of beach. Although the methodology appears sound on reviewing the reports, a number of sub-optimal results were obtained due to flying the actual thermal air survey in the evening instead (2100 hrs) after a long period of aircraft standby and therefore didn't provide the maximum thermal contrast and lacked definition. Similarly, on receipt of the analogue photographic prints there was significant scale distortion and hydrotechnica interpreting the data couldn't accurately overlay OS grid lines onto the prints, there is potential for significant error in grid references as these were attempted using triangulation of landmarks onshore. The locations of the identified zones need to be treated with caution and is a significant limitation of this survey. The results describe that the locations were subject to 'considerable error' but the amount is not determined. Even with the errors and the sub-optimal environmental conditions the survey prints did show several thermal anomalies within the intertidal zone that then warranted further field investigation.

The areas mapped from the thermal survey, and the follow up geochemical survey did not show beach springs in the area previously described in the earlier 1970-1980s beach investigations. However, there was one interesting anomalous sample point north of the Sellafield site, approximately 3000 metres north along the beach from the beach seepages and 500 metres south of the Braystones railway station, showing abnormal tritium levels on both rounds of spring water sampling in April 1993, and March 1994. The levels were 2330 TU and 1280 TU respectively. There were no other high levels along the beach recorded nearer the Sellafield site. These results could indicate that there is tritium making its way from the nuclear site to this area but doesn't align to the region that was recorded in the earlier investigations. It wasn't clear how far north they surveyed up the beach in the late 1970s investigations, is it possible they missed some tritium beach discharge points? There is a small tarn area to the east of this point on the beach, it may be possible that tritium from rainfall in the area might have concentrated in the tarn sediments over time caused by accidental aerial release from the known silo fires, but lake water results in Whittaker (1977) doesn't show any high levels of tritium locally. If the contamination is from the nuclear site, then a groundwater pathway may be linked somewhere within the main sandstone aquifer. No resolution of the origin of this abnormal result is reported.

2.5.5 Further hydrogeological site investigation at Sellafield beach (1995-2009)

Several phases of the Sellafield conceptual model and groundwater flow modelling (Sears, 1994; Nirex 1993; Sears et al., 1994; Sears & Thompson, 2003; Smith & Cooper, 2004; Randall et al., 2004) were developed over the late nineties and a large Sellafield contaminated land and groundwater management project carried out up to 2010 however no specific investigations took place in the river Ehen and beach springs coastal boundary area from 1995-2009, with the exception of a geotechnical investigation of the ground conditions (Garrison, 2009) just south of the sea pipeline for a temporary bridge construction project to move materials from a barge over the river to the Sellafield site, the borehole logs confirmed glacial deposits and sandstone as per the earlier studies, however there were no radionuclide surveys carried out. A new programme of beach springs field measurements was started in 2009 as part of this research project in parallel with the review of the historical documents and the findings are described later in Chapters 3 and 4.

2.6 Long-term Temporal Variation in Tritium Activity in Beach Springs (1976-2009)

Water quality spot samples of tritium activity concentrations for beach springs discharge area described in Section 2.5 have been collated for the period of 1976 to 2009 from the historical documents and the Sellafield Ltd environmental monitoring records 'EAGLE' database and converted to Bequerels/litre. The descriptive statistics for the 299 sample results are presented in Table 2-4, it should be noted that there may be some data gaps from records not found during the search but it is considered that there is sufficient data points (>100) covering seasonal and tidal cycles each year, to give confidence in the trends presented.

Table 2-4: Descriptive statistics of tritium activity (Bql^{-1}) in beach spring discharge area on Sellafield Beach from 1976 to 2009.

	Tritium activity in freshwater component of the Sellafield beach springs (Bql^{-1})				
Year	Number of Samples	Max	Min	Mean	Standard Deviation
1976	10	35890.00	8510.00	24364.50	9664.98
1977	25	40700.00	8880.00	22725.40	8287.86
1978	8	34743.00	10508.00	22884.50	8448.76
1979	12	40774.00	2183.00	18777.50	11239.69
1980	12	32560.00	4995.00	17898.75	7800.95
1981	11	28638.00	3700.00	15940.27	7571.56
1982	11	30118.00	2146.00	11587.73	8100.72
1983	12	25604.00	1961.00	13449.50	7456.50
1984	12	13172.00	1165.50	6596.79	4045.94
1985	12	20350.00	1073.00	8913.92	6082.14
1986	12	20000.00	1500.00	9858.33	4977.12
1987	12	4000.00	230.00	1528.33	1085.83
1988	8	3700.00	520.00	1920.00	1188.74
1989	2	1000.00	470.00	735.00	374.77
1990	7	1500.00	180.00	740.00	632.14
1991	12	1900.00	140.00	688.33	521.36
1992	12	2100.00	160.00	1047.50	584.51
1993	13	1350.00	418.00	813.31	298.32
1994	12	1030.00	220.00	720.42	268.40
1995	10	760.00	120.00	478.40	237.23
1996	9	1430.00	116.00	579.89	407.36
1997	12	961.00	189.00	565.08	219.84
1998	11	558.00	119.00	391.27	153.69
1999	12	970.00	112.00	384.83	247.17
2000	4	298.00	105.00	162.75	90.67
2001	3	619.00	178.00	419.33	223.43
2002	4	554.00	121.00	351.25	178.03
2003	4	809.00	474.00	676.50	156.02
2004	3	1950.90	119.61	732.19	119.61
2005	5	187.06	102.80	128.97	33.45
2006	2	623.49	246.26	434.87	266.74
2007	2	246.22	179.37	212.79	47.27
2008	3	411.56	145.22	305.06	140.95

The time series profile for tritium activity in the beach springs area from 1976 to 2009 has also been reconstructed (Figure 2-10). The plume is thought to have reached the beach between 1952 (earliest record of tritium waste being buried in trenches) and before 1975, and then experienced a steep decline between 1976 and 1990 and has stabilised at low tailing concentrations (below 750 Bql^{-1}). There are secondary peaks of less magnitude in 1982-3, 1986, and 1992 and supports the earlier hypothesis of a multiple plume model, further minor fluctuations are seen on the shoulder of lower concentrations.

In general the trend is characteristic of a decaying tritium source term (half-life) however the smaller scale fluctuations (peaks and troughs) seen in the tritium concentrations over the period may suggest that further smaller pulses of release from the source term(s), perhaps a breakdown of waste wrappings or minor discrete plant leaks events are appearing at the foreshore, which are considerably smaller than the event recorded in the 1970s but the signatures are still picked up in the beach springs. It should be noted that in the early 1990s the site went through a period of capping (with tarmacadam or concrete) of any open run off areas, to reduce rainfall infiltration and recharge. The data may suggest that this in part slowed down the leaching of tritium from the source areas and the plume appears to have stabilised, as a further reduction in tritium levels is recorded post-1990.

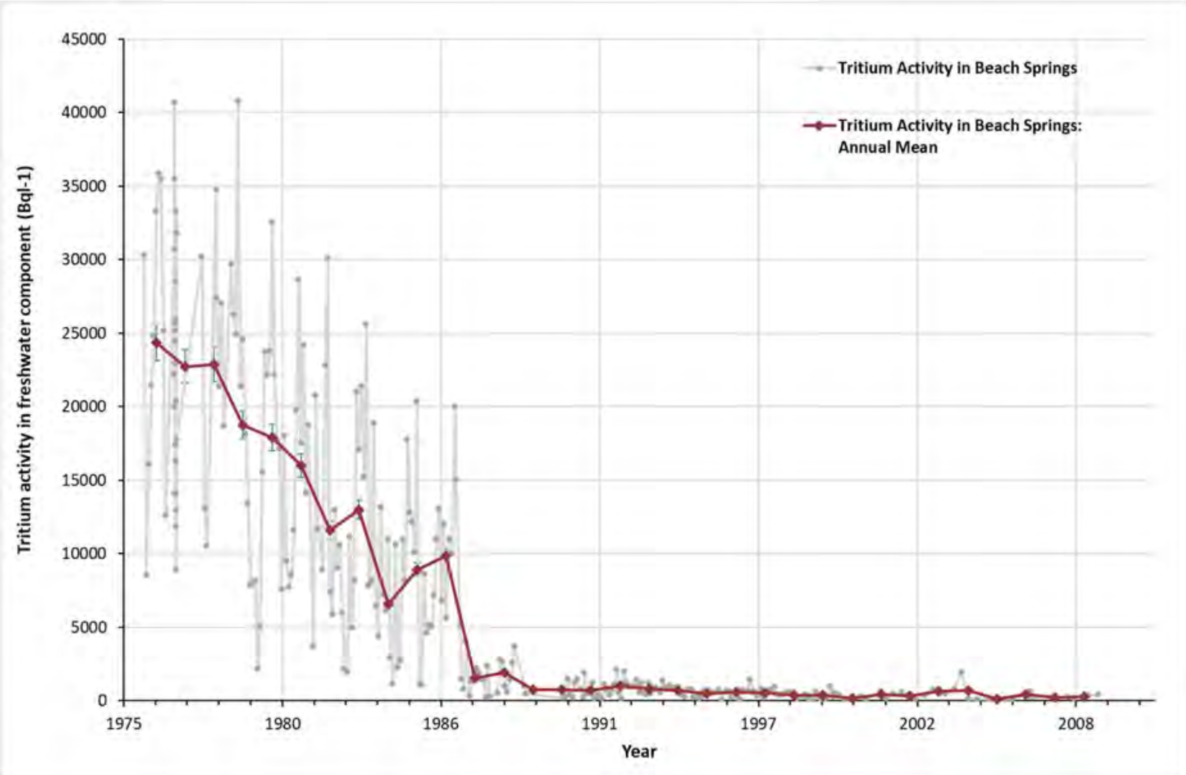


Figure 2-10 Time-series profile of tritium activity in the freshwater component of the beach springs discharge area 1976-2009.

Seasonal fluctuations are apparent in the beach springs activity concentrations during an intense sampling period of 1980 to 1986, and this response to rainfall can be seen in Figure 2-11, where low monthly rainfall levels correlate with higher tritium activity concentrations, due to reduced dilution effects. It is also possible that given the river Ehen is tidally influenced further dilution effects could be applied on cycles where the tide is higher and reaches as far as the pipeline in the river Ehen in the area near the beach springs discharge zone.

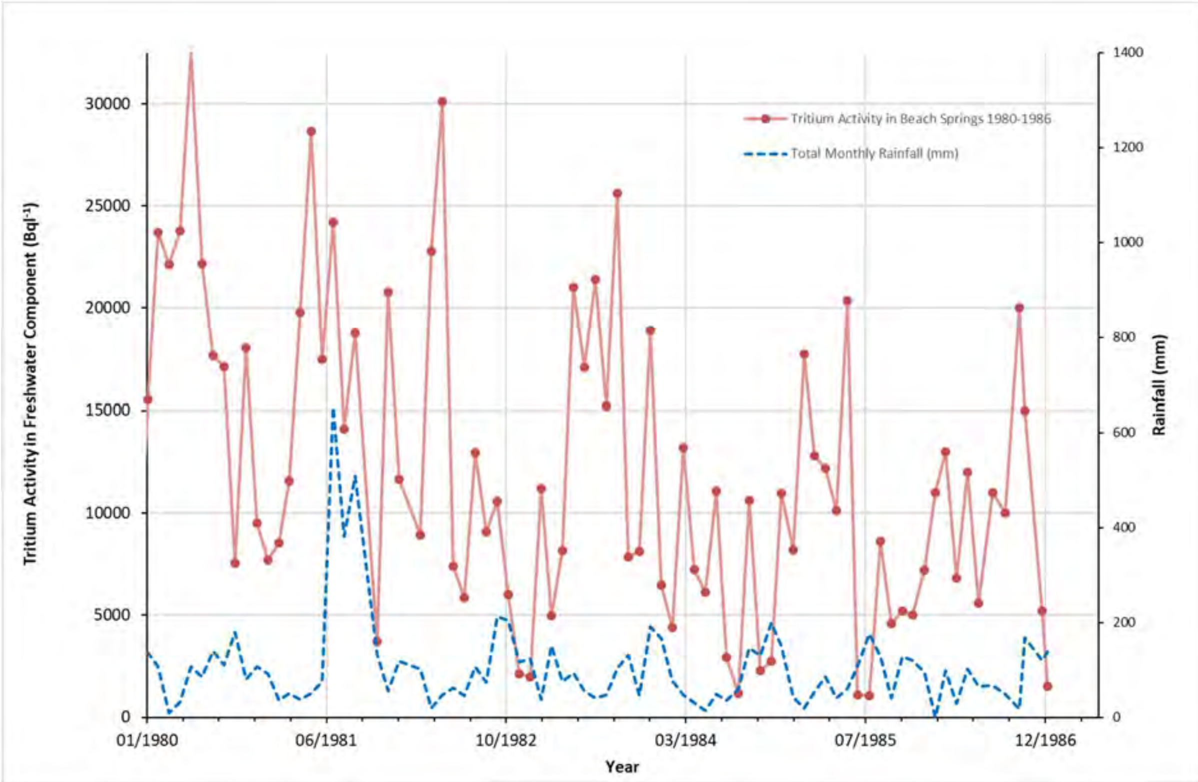


Figure 2-11 Fluctuations in monthly tritium activity concentrations at the beach springs in relation to potential dilution effects by rainfall recharge (1980-1986).

2.7 Tritium Activity in Beach Area Groundwater Monitoring Wells

The beach groundwater monitoring boreholes installed in the 1970s either side of the river Ehen were water quality bailer sampled and tritium activity annual means are presented in Figure 2-12 and support the temporal tritium trend seen in the beach springs discharge. There is a gap in the data in the mid-1990s although interpolation of the trends looks simple. Monitoring well BHP03 and BHP47 are nearest to the beach discharge zone, and the secondary peak around 1986 is seen clearly in their time-series data, further supporting the multiple plume theory and likely linked to the plant leaks described in Sections 2.4 and 2.5

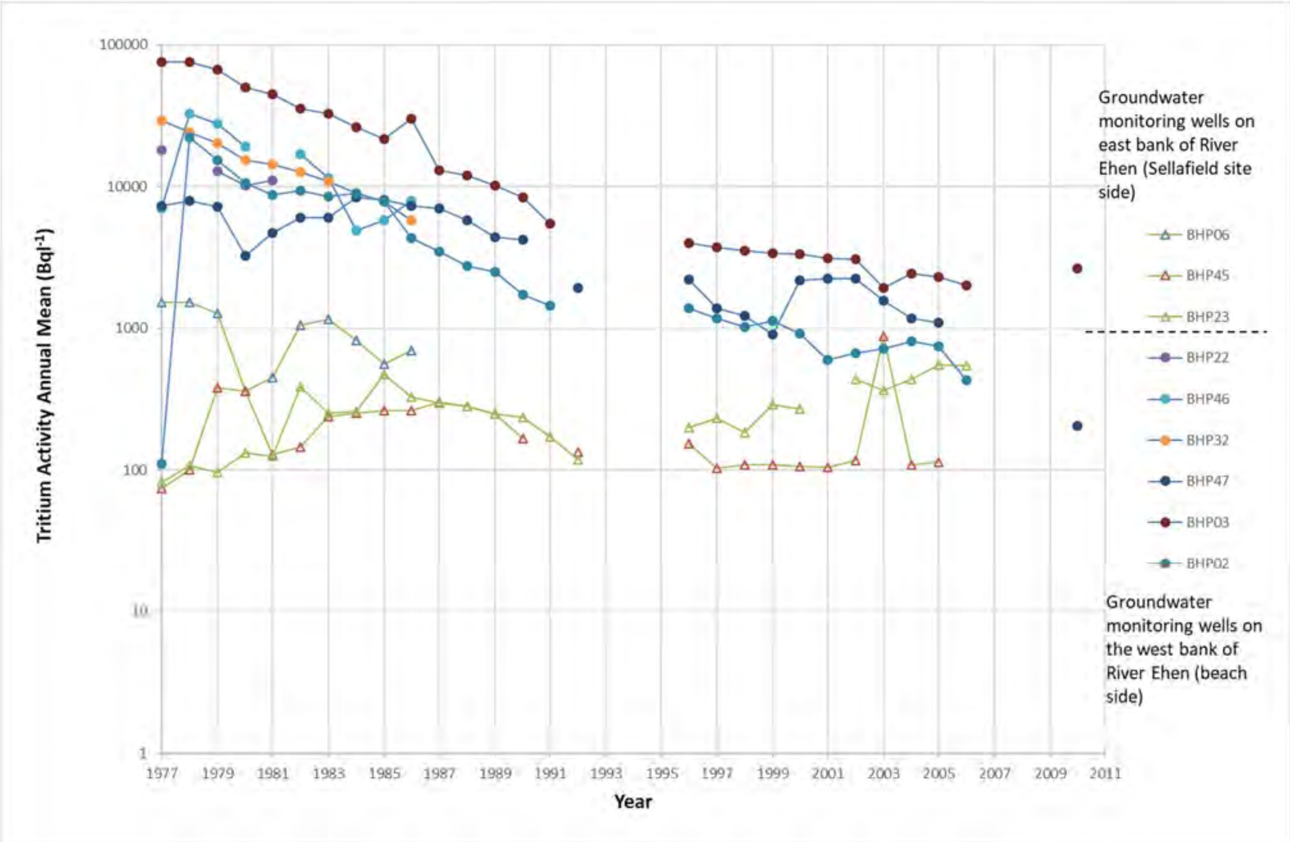


Figure 2-12 Annual mean tritium activity time-series for groundwater monitoring wells located either side of the river Ehen during the early beach investigations (1977-2010).

The groundwater monitoring wells on the east side of the river Ehen, on the Sellafield site perimeter side show lower concentrations of tritium for the full period, in the range of 100-1500 Bql^{-1} unlike the beach springs and groundwater monitoring well tritium concentrations on the west side of river, suggesting that groundwater pathways are complex at the river boundary. Further investigation into the interactions between the river Ehen and the groundwater are required to better understand the flow dynamics in this area.

A north-south cross section of the tritium activity on the west (beach) side of the river Ehen groundwater monitoring wells indicate that there are multiple peaks of the plume centred around the groundwater monitoring wells BHP03 and BHP32 and are presented in Figure 2-13 as annual profiles. It can be interpreted that the tritium is forming narrow plume peaks and merging at the river/coastal boundary and expressed in this foreshore zone both in groundwater wells and the beach springs.

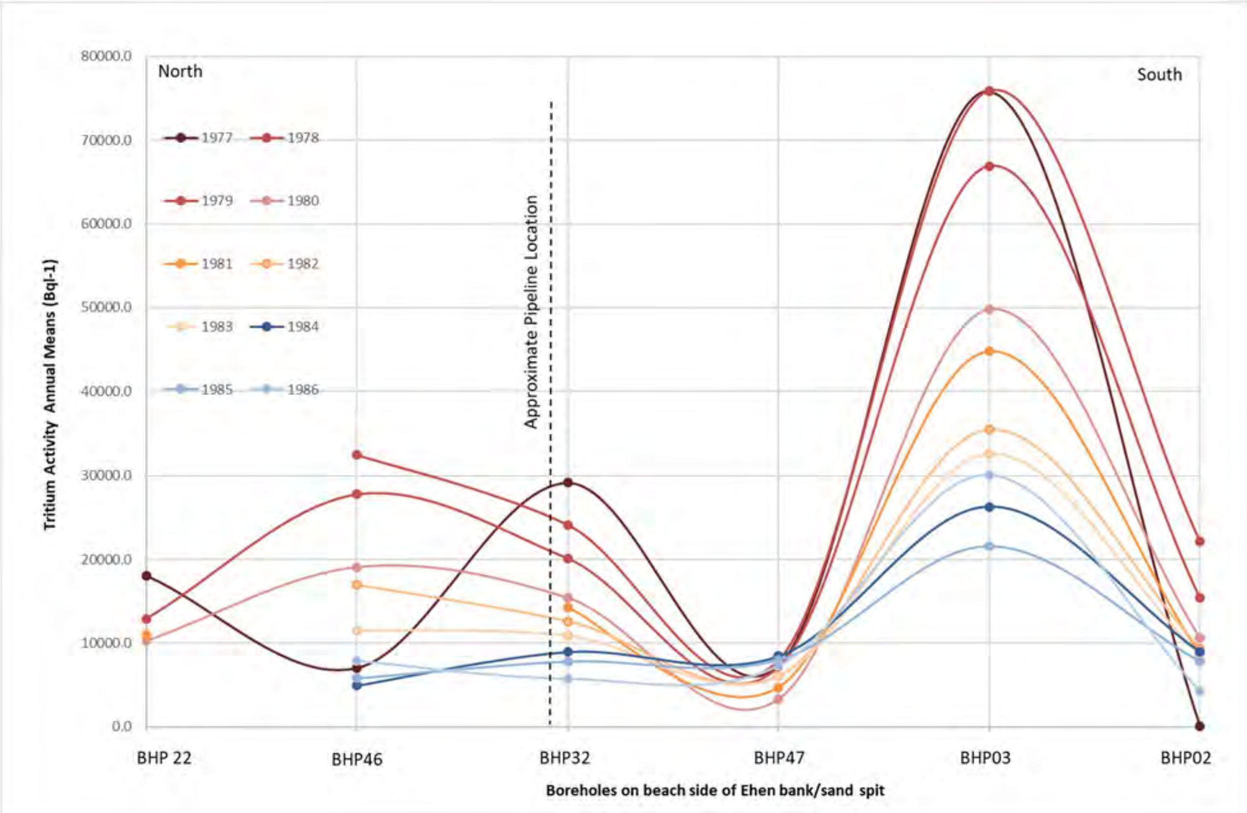


Figure 2-13 : N-S cross section of annual mean tritium activity levels in the beach groundwater monitoring wells (BHP22,46,32,47,03,02) along the west side of the river Ehen along the beach sand spit area.

Many of the early 1970s groundwater monitoring wells were retired (depending on condition) or sampled less frequently as the newer series of groundwater monitoring wells were installed and came online in the growing network over the last four decades at the site. More modern groundwater monitoring wells were drilled on the west side of the river Ehen along the sand spit in the early 2000's (known as the '69' series) and their locations are either side of the main beach springs discharge area and should be reviewed for more recent trends. The tritium data from the full groundwater monitoring network should also be reviewed to identify any further indicators of further tritium fluctuations/peaks to arrive at the beach springs. Confidence that no further tritium peaks are expected would support confirmation of the observations of this review that the historical tritium plumes are stabilised and at low risk tailing concentrations.

2.8 Synopsis: Preliminary Conceptual Model for Beach Springs Area

The assimilation of the geological, hydrogeology, pathway and discharge information from this review can be interpreted into a preliminary conceptual model for the beach springs discharge zone, and cartoon schematics are presented in Figure 2-14 and Figure 2-15, interpretation of the data throughout this review is incorporated with the original conceptualisations presented in the archived reports particularly Whittaker (1977;1978), Holmes and Hall (1980), Smith (1982) and Oakes (1985). The estimated location of the buried channel is adjusted from the original interpretation following review of the geophysics information and the geological interpretative report from the more recent on-site contaminated land characterisation project (Garrick, 2010) and estimated location of faulting from review paper by Cooper (2009).

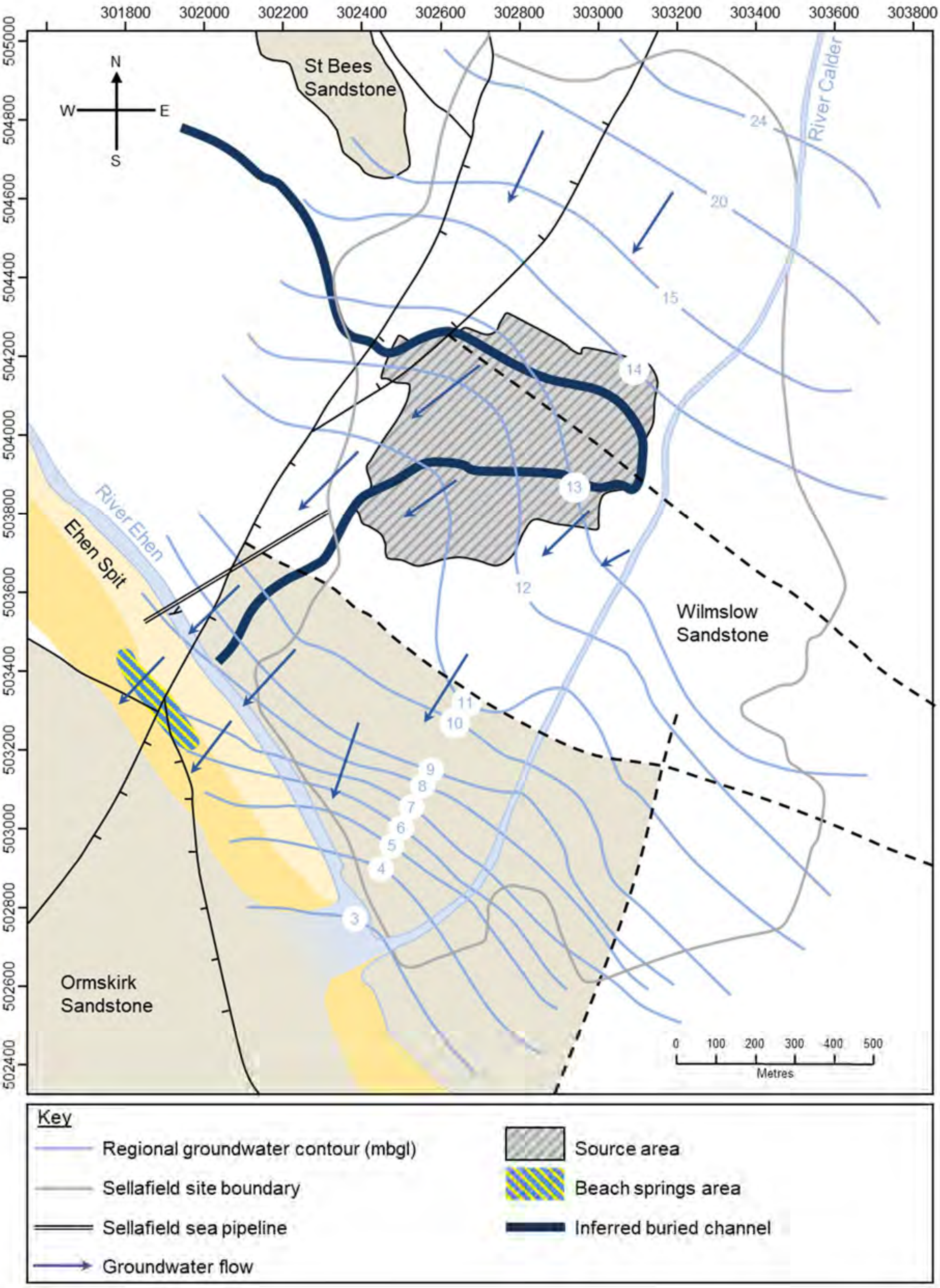


Figure 2-14 Schematic conceptual plan view of source area and groundwater flow towards the beach springs area collated and interpretation developed from historical archive review (Whittaker, 1978; Holmes & Hall, 1980; Marples, 1980(a); Smith 1982; Williams, 1984; O Oakes, 1985(a); Cooper 2009; Garrick, 2010).

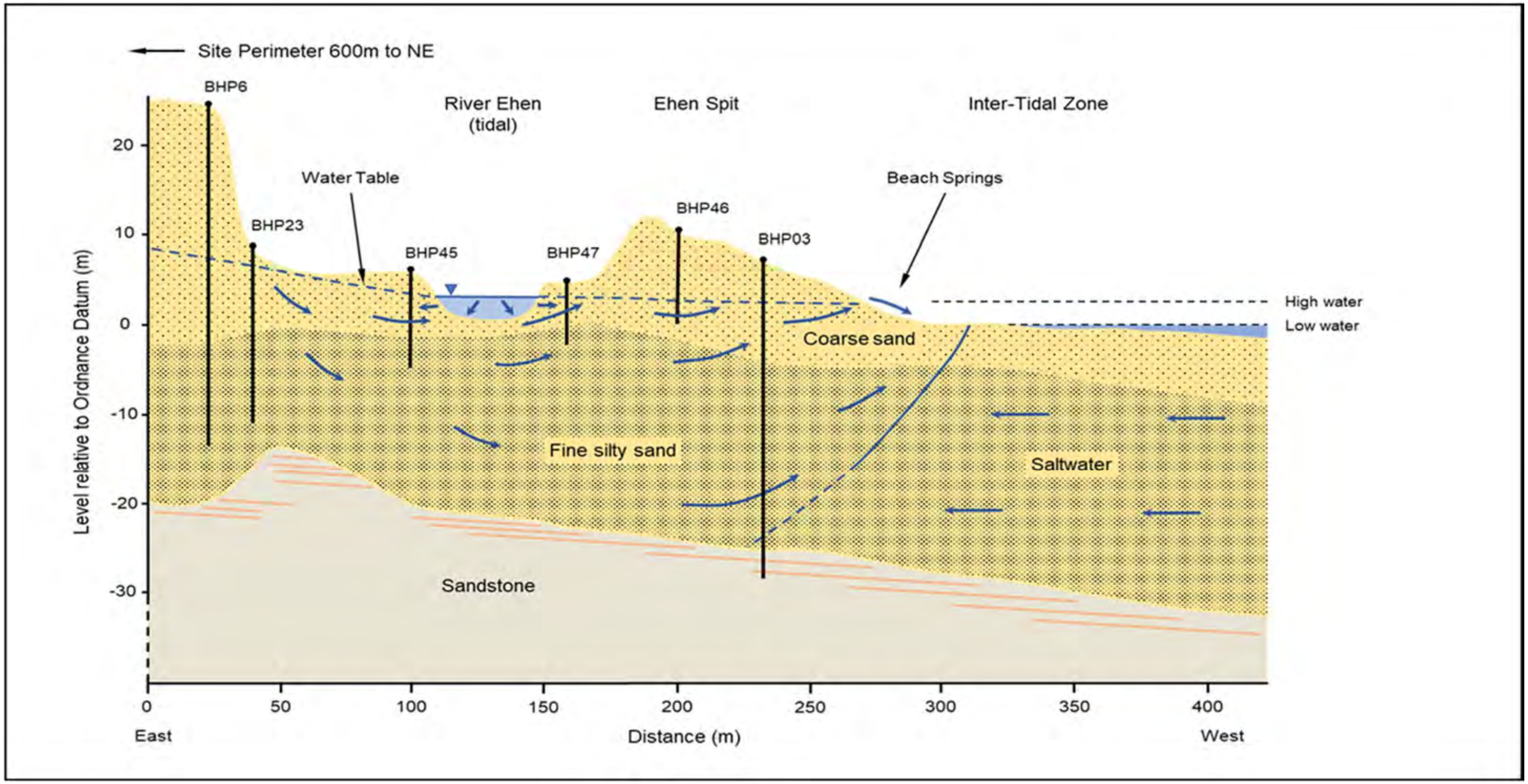


Figure 2-15 Conceptual cross section of groundwater contamination pathways from the Sellafield site to the beach springs discharge zone collated and interpretation developed from the historical archive review (Whittaker, 1977; Holmes & Hall, 1980; Marples 1980(a); Smith, 1982; Williams, 1984; Oakes, 1985(a))

There a range of different likely source term contributors but there remains some uncertainty throughout the individual documents, the review carried out during this study of the full timeline of evidence and information points strongly to the Windscales disposal trenches as the primary dominant source term that caused the peak levels of tritium at the beach springs prior to 1975, with other narrow plumes of less magnitude also migrating to the beach. Groundwater flow pathways with tritium fingerprints from multiple sources appear to be merging into a front as the groundwater moves towards the coast and the expression of those combined flows is seen at the beach springs discharges in the intertidal zone. Early groundwater modelling gave some interesting insights by suggesting a two narrow plume model one that started in the late 1950s following waste emplacement in the trenches, and then an additional plume related to identified leaks in 1978 and 1984, which appears reasonably accurate against the river and beach time-series profiles. Looking at the evidence and number of potential tritium sources there may well be up to 4 or 5 individual tritium plumes (Figure 2-16) tracking with a relatively narrow plume orientations towards the coastal area and expressed in the seepage front in the beach area. It would be important to establish if there was any other contamination seen further north of the spring peak discharge zone noting the anomalous tritium results found 3000 metres north of the site during the aerial beach spring surveys in 1993 and 1994.

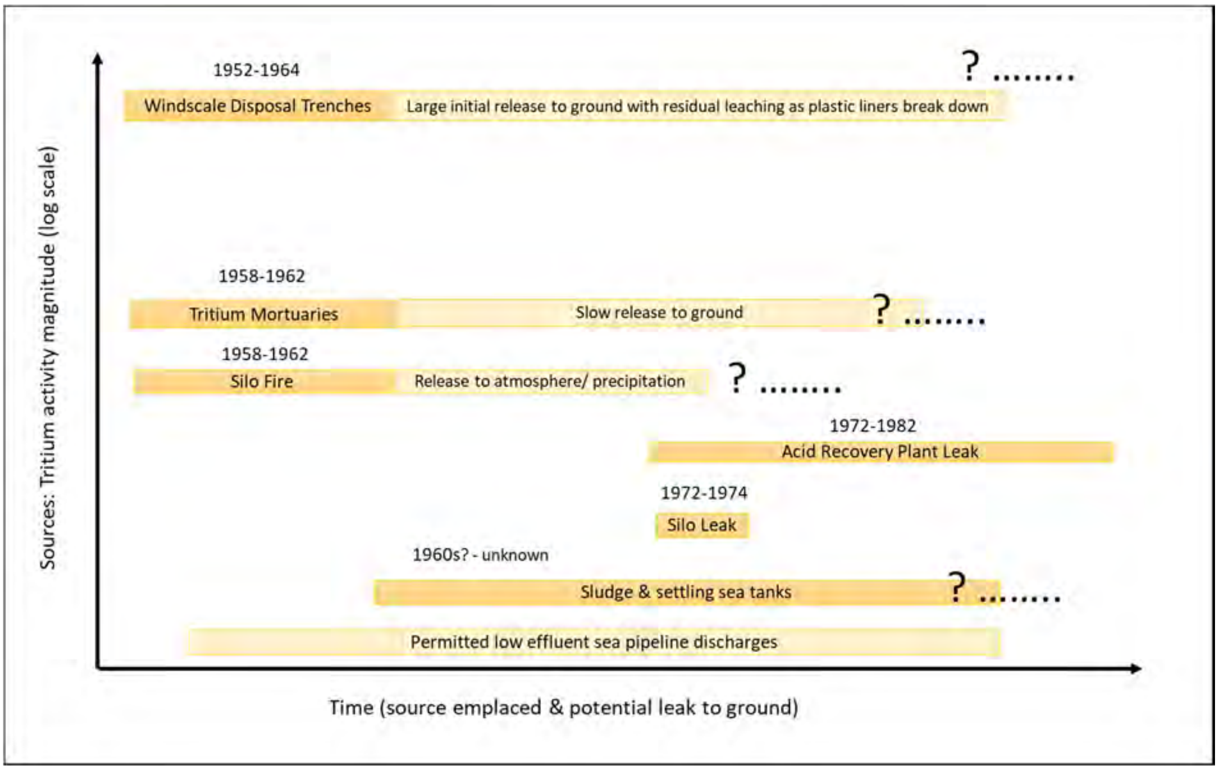


Figure 2-16 Timeline of potential tritium sources that could have entered groundwater during early operations at the Sellafield Site.

2.9 Conclusions

A systematic review was undertaken to assess evidence for a tritium groundwater plume surfacing on a beach near the UK's oldest nuclear site. An initial conceptualisation of the evolution of a tritium plume related to multiple contributing source terms, by merging groundwater flow paths near the coastal intertidal zone (receptor) and surfacing in the intertidal zone in beach springs is presented based on all the information collated and interpreted during the review. Considerable material used in this review remains unpublished, and likely holds historical significance as one of the earliest coastal groundwater contaminant plume investigations in the UK, and possibly one of the earliest inland to be characterised along with a few landfill plumes in the 1980s.

The following conclusions are drawn against the objectives outlined in section 2.1:

(1) Published evidence for the occurrence of radionuclides in beach springs is very limited and there appears an absence of publications to date that record tritium in beach springs in coastal environments in the UK or worldwide.

(2) A search strategy to include grey literature has proved worthwhile resulting in the research and collation of data to frame the research direction to confirm the understanding prior any fieldwork commencement, and hitherto became a rather historical archive of data with much intense activity in the 1970s-80s (with significant details gathered that are potentially forgotten now), but with investigation activity tailing off into the 1990s-2000s where monitoring had become routine and limited to Environmental Permit compliance.

(3) The quality of the collated datasets is assessed as overall moderate to high. The studies were carried out by professional scientists often in association with the Institute of Geological Sciences or similar government scientific bodies.

(4) Evidence provided as part of this systematic review constrains the arrival of tritium at the beach to have occurred after 1952 and before 1975 (earliest date the source could have been emplaced and when the tritium was discovered). There is evidence that there is a several sources of tritium that could have contributed to the plume(s) surfacing in beach springs. The primary contributor being the Windscale trenches and sea tank area, a second more northerly area containing tritium mortuaries and the acid recovery plant and lastly a grouping of minor historical leaks from legacy silos or effluent plants.

(5) The data collated shows that the main peak of the tritium plume arrived at the beach, before it was discovered in 1975 where peak rates are seen by the end of the 1970s and has been in significant decline since this time and now appears to be stabilised at low levels providing reasonable confidence of future low risk levels for the site environmental safety case (SWESC) for tritium, but not necessarily for other (retarded) radionuclides following a similar pathway to this receptor.

(6) The main source area of the Sellafield site may be geologically connected with the beach spring discharges by a buried channel feature that could provide a preferential groundwater pathway, the axis of which also follows the sea pipeline trench route. The significance of the buried channel feature as a preferential pathway for contaminant migration and delineation of its 3-D extent remains an ongoing priority for the Sellafield site.

(7) Although data can be extracted and provide context to the timing of the tritium arrival at the beach, the characteristics of the plume is hard to ascertain from the current data set due to lack of accurate position data, and no repeatable methodology in terms of the type of field measurements taken and minor uncertainties in the laboratory analysis that was carried out. Even with these issues part of the breakthrough curve for tritium at the beach springs can be reconstructed, and this is supported by groundwater monitoring well data and a river-bed survey in the Sellafield beach area.

(8) The groundwater monitoring on the site is focused around the source areas with monitoring existing but typically sparser, towards the site periphery approaching the coast. An outcome of this, as is apparent from the review, is that it is challenging to define groundwater plume pathways and categorically connect sources to the River Ehen and the beach spring discharges.

(9) Monitoring of the contamination expression in the River Ehen riverbed only received investigation in the early years of investigation (1977-1979) and its expression since can only be gauged by the concentration monitoring of the river itself. The early study points to a complex plume discharge with potentially the same plume able to discharge to both the river Ehen and the beach springs and reversal of flows (due to tides occurring) and a possible complex relationship of flow and contaminant exchange between the river and aquifer towards the beach, investigation of this complexity (although recommended in the 1970s-80s) was not undertaken. Early measurements suggest the tritium discharge to the beach and the

riverbed were similar and has not since been investigated and was based on limited late 1970s data.

(10) Monitoring in recent decades have been focused upon concentrations with little attempt to consider contaminant mass fluxes, some of the earlier studies provide valuable flux data that have since been underused.

(11) Beach spring monitoring has historically focused along a relatively short length of foreshore at the historical plume hotspot; it is hence unclear if other discharges occur along the wider foreshore, and if there are further submarine discharges.

It is recommended that further fieldwork will be required to further characterise the tritium plume footprint at the beach springs and improve understanding of the overall system dynamics. The review however does act as a historic baseline comparison for any new data collected and provide a valuable body of summarised research for the future regulatory requirements for a Sellafield site wide environmental case (SWESC).

2.10 Recommendations for Further Work

(1) It is recommended that a field investigation project is initiated to determine whether tritiated groundwater is surfacing in the beach springs at the intertidal zone in the present day (2009 onwards) and if found, use of geo-referenced spot samples to characterise further the spatial extent of tritium occurrence in beach springs on the Sellafield beach (Chapter 3).

(2) The Sellafield site has developed an extensive groundwater monitoring well network over the years and tritium activity data in wells further into the site between the source area and the beach should also be reviewed to understand present day status of migrating tritium, noting that some of the original beach monitoring wells are now no longer in operation. (Chapter 5).

(3) The complex flow interactions between the river Ehen, tidal reach, the beach springs (foreshore) and the tritiated groundwater plume should be explored further (Chapter 3).

(4) Consider simple 1D contaminant transport modelling to test pathway transport times and parameter sensitivities for tritium breakthrough at the beach springs calibrated using the source term information from this review and the receptor tritium time-series field data at the beach monitoring wells and beach springs (Chapter 5).

(5) Consider whether there are any other radionuclides have reached the beach discharge zone in order to further refine the beach springs conceptual model and inform future environmental risks to the coastal area (Chapter 4).

Supplementary Data Appendix:

A2.1: Systematic review spreadsheet for archive research study.

CHAPTER 3 DELINEATION OF A TRITIUM PLUME DISCHARGE FOOTPRINT BY BEACH SPRING MONITORING

3.1 Introduction

The Office for Nuclear Regulation and the Environment Agency jointly regulate contaminated land and groundwater on the Sellafield site via a Joint Memorandum of Understanding. The Sellafield Environmental Monitoring Programme is a requirement of the Environmental Permit (KP3690SX) that addresses the disposal and monitoring of discharges of radioactive material to the environment. Further to this is an additional permit the Sellafield Environmental Permit for Pollution, Prevention & Control (BM4317X), which is related to the use and disposal of non-radioactive materials and includes groundwater monitoring of non-radioactive chemicals of concern (Sellafield Ltd, 2016a).

The Sellafield site has a significant groundwater monitoring well network infrastructure and the Sellafield conceptual model has been built from over four decades of groundwater water quality data. The network and monitoring regime have continuously evolved since the discovery of tritium in the groundwater and beach springs in the 1970s. The literature described in the previous chapter details field investigations in the beach springs area near the Sellafield site carried out predominantly in the 1970s-1980s and a preliminary conceptualisation for tritiated groundwater surfacing on the beach was presented following analysis of the archived investigation reports.

There are about 500 active groundwater monitoring wells available, subsets of which are used in the annual groundwater programme investigations. For example, in 2016, an array of 181 of the monitoring wells (typically with 2m or 4m length screens) was used for both groundwater quality low-flow sampling and level monitoring with around 700 samples taken for radionuclide and supporting hydro-chemical analysis (Sellafield, 2016a). Additionally, monitoring of local surface waters, seawater and beach springs is routinely undertaken (quarterly) to determine whether environmental radionuclide levels remain within acceptable levels of the permit and below acceptable public risk levels. Despite the significant monitoring at the site, the spatial extent of the plume discharge area remains poorly defined at the shoreline. Also, the contributing source and plume areas on site and connecting pathways remain subject to some uncertainty and debate. This is despite the site representing one of the UK's most characterised and monitored sites. The delineation and the estimation of contaminant plumes in groundwater systems at the field scale still remains a challenging task for all types of industrial areas (Datta et al., 2016).

A major reason for the poor definition of the discharge area is that a greater proportion of groundwater monitoring wells are in and around potential source terms with a mandate in part to contribute to 'leak detection' monitoring. The source areas are well characterised and plumes in and around those facility areas. However, the possible numbers of sources, the complexity of the superficial deposits with heterogeneity and the buried channel, along with the accompanying deep sandstone aquifer that is faulted, a complex flow regime (two rivers, coastal interface and on-site water leakage) makes for a complex proposition for precisely defining plume migrations. Whilst the wider site area migrating contaminant pathways are reasonably monitored (but less than source areas typically) allowing fair migrating plume spatial definitions and informed predictions of risks to receptors, the spatial resolution of detail in the mid to far site areas and nearing the beach-river receptors that allows precise resolution of plume detail and trajectories is still lacking. Moreover, at these more remote locations there is increased potential for merging of plumes and their contributing sources terms at distance become less clear.

This chapter therefore presents data on spatial and temporal variations in tritium beach discharge and delineates a groundwater tritium plume at the foreshore. The aim is to use the geospatial analysis of the beach spring discharges to provide a new perspective on the tritium plume characteristics, and therefore develop further the conceptual model of the system that may underpin management of risks posed. The objectives of the field investigation were: (1) obtain field data characterising the nature and significance of the tritium groundwater plume beach discharge footprint over the decade 2009-2018; (2) to compare the observed modern discharges with those indicated by the archive beach spring discharge and monitoring well data spanning some four decades; (3) to construct long-term temporal trends; (4) to outline possible controls upon the tritium discharge footprint; and (5) to consider how the observed footprint informs future risks posed from tritium and more attenuated radionuclides that may have later arrival times.

3.2 Materials and Methods

3.2.1 Beach spring monitoring and sampling framework

3.2.1.1 Survey design

A initial field programme of beach spring monitoring and geochemical water quality sampling started on the 15th June 2009 and took place for six months covering both the summer and winter season on a fortnightly basis, starting to collect data and samples at the lowest point of the tide to maximise spring flow and maintain a safe working window in the tidal zone. Freshwater discharge is correlated positively to low tide level, which infers that when low tide elevation is higher during the neap cycle more freshwater discharges as a proportion of total discharge and when low tidal elevation is at its lowest, discharges are expected to be more saline (Abarca et al., 2013).

A sampling procedure was written and included a remote coastal environmental risk assessment and the sampling methodology and analysis plan for each field visit. The remote coastal environmental risk assessment included: access; egress; 4x4 vehicle and water rescue training; dangerous wildlife assessment; protection of ecology by wheel washing; maximum safe operating window – 4 hours from lowest tide; no lone working; security and safety protocols for working near a nuclear site; land owner permissions; transfer of samples; chain of custody arrangements; and emergency arrangements.

Cognisant of the historical archive of beach spring reporting for the site, the study area was selected to monitor a 3.5km section of the beach from the Sellafield site northwards to the Braystones railway station (Figure 2-1) to ensure that the spatial extent of any tritium activity was captured, noting the anomalous tritium results found further north of site in 1993-1994 (Section 2.5.4). There was no evidence of tritium on the beach south of the Sellafield site from previous environmental monitoring, and groundwater monitoring well data indicated very little radiological contamination on the south side of the River Calder and therefore this area was excluded from the study. The spatial frequency of sampling at each field visit was determined by sampling from all visible beach springs. These often congregated into small channel flows on the beach (Figure 3-1). The monitoring study section length was subsequently reduced to approximately 2.0-2.5 km as the data started to reveal the spatial extent of the tritium footprint. Initial sampling frequency was fortnightly, but this was reduced to quarterly from February 2010 to September 2018 to balance economic and resource constraints but still retain good annual coverage over the winter and summer months.

Figure 3-1 Photograph of the beach spring discharge zone during low tide conditions. Sampling along the visible seepages at the edge of the gravel line shown.

3.2.1.2 Event Sampling approach

All beach spring measurements and collection of water samples were carried out at low tide on the same day for each collection round. Sampling started at the south end of the Ehen Spit and worked northwards. Springs were visually identified where water was seen at the beach seepage face. GPS location data was recorded for each sample. Specific electrical conductance (SPC) normalised to 20^{o}C in-situ measurements were taken with a single parameter handheld device during 2009 and then the meter was upgraded to allow multiple field parameters to be recorded from 2010 onwards. From 2010 SPC, pH and temperature measurements of the spring water were recorded in the field using a YSI multiparameter handheld water quality meter. The handheld meter was calibrated with the manufacturers (YSI) standard solution prior to each data collection round. The samples were only collected from areas that measured a SPC level of 45 $mScm^{-1}$ or less to indicate that the waters contained a proportion of groundwater, rather than seawater runoff whilst the tide retreats (Rusydi, 2018). On two occasions (27/08/09 & 09/11/09) blank, split, and local surface water samples from rivers and tarns near the beach were collected. The samples were collected over the course of the low tide window at different times of the day from various locations this meant that samples at specific geographic locations were taken at different times of the tidal cycle. As such their expected proportion of freshwater versus seawater may vary. To minimise such composition differences between samples taken on different dates from a given geographical locality, the same sequence order of site sampling was followed each sampling mission allowing retrieval at comparable times in the tide cycle at a given locality. Twice the sampling direction was reversed (12/10/09 & 26/09/14) along the beach from Braystones (north) to near the site (south) and the sample results were within 10% of the usual sequence order.

3.2.2 Tritium sampling and analysis

Water quality samples were collected routinely from June 2009 until September 2018 at low tide from the beach springs at the foreshore. An unfiltered one litre bulk spring water sample for radiological testing was collected at the beach spring, as close to its exit point on the beach as possible. The sample container was rinsed with deionised water, and then rinsed with water from the spring to ensure no cross-contamination issues and then over filled (to remove air), sealed and transported to the local UKAS accredited laboratory at Westlakes Science Park, Cumbria.

Tritium was assayed by liquid scintillation counting and checks were made for quenching and/or contamination by other emitters (QAAM 99, 2014; ISO 2015). A suitable portion was made alkaline with sodium hydroxide. A 20 ml portion of the resulting alkaline solution was transferred to a 9cm diameter polystyrene petri dish and warmed on a hot plate for 20 to 30 minutes at 50 to 70˚C. A 1ml aliquot of the condensate was transferred from the lid of the petri dish to scintillator solution. The

Tritium data were decay corrected from the time of sampling to the time of analysis by the radioactive half-life of 12.32 years, and all tritium concentrations presented are expressed in the standard unit of Bql^{-1} and represent the sample activity concentration of tritium found in the sample at the beach face. The limit of detection of tritium analysis for the 2009-2018 samples was determined by the minimum detectable activity (MDA) of 7.5 Bql^{-1}. MDAs have varied over the last forty years of monitoring and earlier samples (pre-1990) were in the range of 20 – 250 Bql^{-1} and improvement in MDA to the 7.5 Bql^{-1} was in 2004. Filtering of samples was carried out at the laboratory to determine seawater % of the sample and then a calculation to determine the tritium freshwater component of the spring waters based on percentage of seawater in a sample filtrate, the freshwater component was calculated using the decay corrected result as follows:

$$\textit{Freshwater component} = 100 \times \textit{Result} - (\%\textit{Seawater} \times \textit{filtrate})/(100 - \%\textit{seawater}) \qquad \text{[equation 3-1]}$$

If calculated result is <MDA, filtrate calculated result <MDA or calculated result is < (% seawater ×filtrate), the freshwater component result is set to not determined (taken from QAAM 76, 2013). It should be noted that sample tritium is deliberately presented alongside the calculated freshwater component results, there was some concerns that the methods for this calculated result may require review by the laboratory and Sellafield Ltd, in some rounds very high seawater results were being obtained yet the SPC data was lower in the field.

3.2.3 Additional geochemical analysis of beach spring waters

Additional geochemical analysis was carried out in the peak discharge spring area on the 9th February 2010 to understand if there were any results that might provide key markers that could be used to fingerprint the potential contributing source area. The analysis included TDS, Beta activity, Mg, Ca, Na, K, HCO_3, Cl, SO_4, Fl, ^{99}Tc, ^{3}H, ^{7}Be, ^{40}K, ^{137}Cs, Alpha, NH_{4+} and NO_3. The sample analysis was unable to rule out any sources based on the chemistry observed and this line of investigation was not taken any further in this study. The beach spring water quality samples collected from 2009 to 2018 were also analysed for technetium-99 and the results are presented in Chapter 4.

3.2.4 Beach spring response to tidal effects

To gain better understanding of any response of the beach springs to tidal influences an experiment was carried out on the 8th August, 2018 to monitor one of the prominent beach spring flow rates and tritium concentrations from the point of low tide, until the beach springs were submerged as the tide progressed up the foreshore. This aimed to reconstruct a similar experiment carried out on the 17th January 1977, where variation in tritium concentrations were recorded between tides (Whittaker, 1977). Water samples were collected hourly for tritium analysis (method described in section 3.2.2), and beach spring flow rates were measured in hourly intervals. This was achieved by capturing the flow using a small V notch weir dug into the sand. A stop watch and 1 litre vessel were then used to sample and measure flow rates, the measurement being carried out three times and averaged. A comparison of SPC, temperature, pH, and tritium has also been carried out at hourly intervals against the tidal phase (neap or spring cycle) during this experiment. Tidal data was obtained from the UK National Oceanography Centre, from the NERC funded anyTide app.

Hibbert (2017) carried out a similar short field study, over five days in August 2017 to record discharges of beach springs on Sellafield beach during low tide using two different methods, a V notch weir and a float channel method. He also attempted to measure head in response to the falling tide.

3.2.5 Groundwater monitoring well tritium data analysis

Tritium laboratory analysis methodology for groundwater monitoring well samples is the same as for beach spring water samples outlined in section 3.2.2. There is a large amount of groundwater monitoring well data available for the Sellafield Site. There are number of boreholes (BH6975; BH703; BH747; BH702; BH6911; BH6978; BH6977) that are situated along the Ehen spit area that run parallel to the beach-foreshore and in closest proximity to the beach springs discharge zone, and others that are on the east bank site side of the river (BH 745; BH6919).

A bailer water sampling technique was used for BH703; BH747; BH745 and BH702 as these are an older series of wells that were installed in the 1970s, and there headworks were installed for this method and low flow purge sampling is used for the more modern wells that have a multi piezometer set up and started operation in the 1990s (BH6975; BH6911; BH6978; BH6977: BH6919). The low flow purge sampling technique does provide more accurate results due to the ability to stabilise flow before measurements are taken, although the bailer technique is still a valid technique for collecting water samples for tritium analysis.

New tritium data from 2010 onwards, have been added to the beach borehole temporal profile described in the previous chapter (Section 2.7) and a further review of the tritium activity in the wider site groundwater monitoring network is presented in Chapter 5 to identify any pending tritium contaminant plumes yet to reach the beach area.

3.2.6 Interactions between the river Ehen, the beach springs and the sea

In order to determine and understand the flow mechanisms between the tritiated groundwater and the tidally influenced river Ehen, a number of field studies were originally planned at the outset of the study: (1) collect surface 'grab' samples from exposed river bed or bank sediments and analyse them for tritium, technetium-99 and strontium-90, the known more mobile species in the groundwater at the site (2) install drive point mini piezometers (Rivett et al., 2008) into the river-bed to monitor the hyporheic zone; (3) collect and analysis river-bed core samples using a hand auger technique (to a depth of 1 metre); and (4) install dynamic density data loggers (AquaTroll 200©) with vented cables to monitor water levels, SPC, salinity, total dissolved solids and temperature in beach groundwater monitoring wells either side of the river Ehen. The investigation work was inevitably limited to periods when tidal and weather conditions allowed access to the river.

On the 4th and 5th of April 2013 the river level was low enough to access after a period of very dry weather (more than 3 weeks of zero rainfall) to carry out a preliminary field visit. The river Ehen typically has a level range of 1.4 metres and mean flow of 5.339 m3/s (1974-2018) as recorded in the National River Flow Archive (2019). Surface grab river-bed samples were obtained using a trowel and sent to the laboratory to separate the liquid from the solids and carry out further geochemical analysis (Figure 3-2).

Attempts to retrieve core samples with a hand auger failed because the riverbed was too consolidated, a mix of dense sand and gravels. The field visits over the previous months also raised concerns that the flow rates and rapid river level changes from rainfall and tidal fluctuations may be too challenging for mini-piezometers installation and a more substantial or flexible installation may be required. Consequently, the proposed drive point mini-piezometer installation had to then be abandoned. However, it is still recommended that a survey of the hyporheic zone should be carried out but with mechanically emplaced piezometers. These would contain pressure and EC loggers and be installed in a transect across the river. An alternative might be to explore other appropriate new technologies such as freeze coring (Freitas et al., 2015) or distributed temperature sensing (Krause et al., 2014), but high river flow rates and tidal influence may limit what is possible.

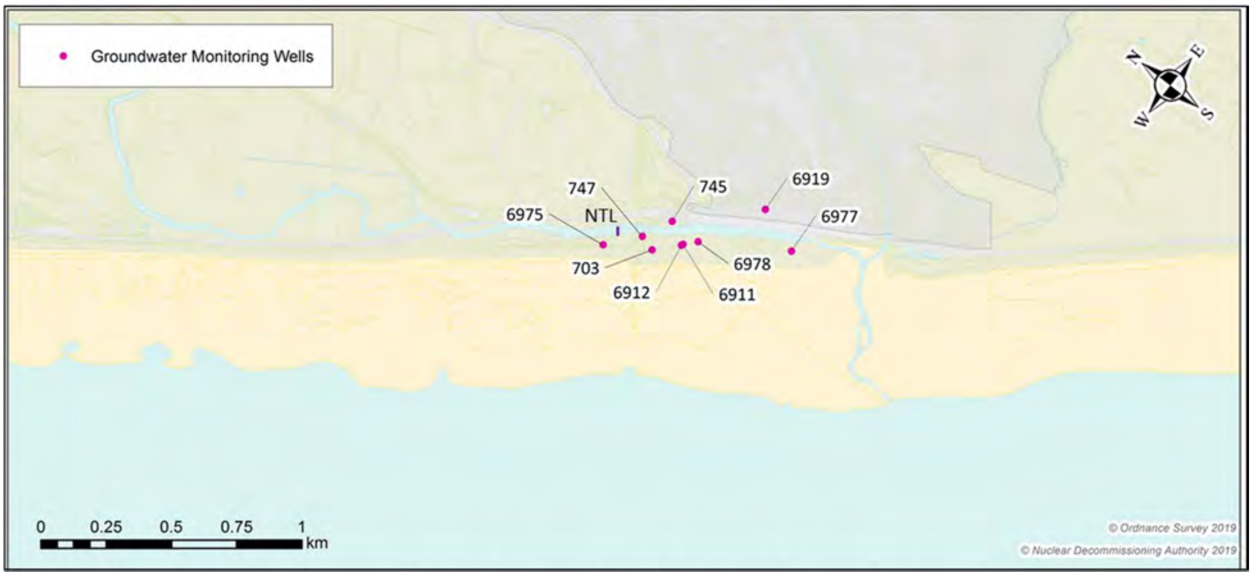
Figure 3-2 Photographs of the field visit to investigate the river-bed composition at very low water levels a) river Ehen looking south down towards the sea; b) river Ehen looking north towards the pipeline; c) consolidated gravel bed, grab samples attempted; d) example of river bank sediment grab sample; e) consolidated large gravel at the surface and subsurface, auger would not penetrate; f) sandy river bank sediment grab sample methods

In addition, in situ monitoring technology for tritium is not yet developed and would still require discrete sampling from piezometers, and the same is true for other radionuclides such as ^{99}Tc and ^{90}Sr. The results of such investigations should be included in the Sellafield conceptual model as it is important to understand the natural attenuation properties of this key groundwater pathway (EA, 2009; Sophocleous, 2002).

The installation of the data loggers into the beach groundwater monitoring wells was successful. They were set to record every 15 minutes to monitor any potential tidal influences. Figure 3-3 provides the locations and set-up parameters of the loggers in the coastal monitoring wells. The loggers were selected due to their ability to manage the change in density should saline water encroach in the groundwater (calculates the head relative to the actual density in real time), important in a tidally influenced coastal area and due to the limits of the budget the loggers had to be moved between wells to get wider coverage. The loggers were then recovered and used in a well transect that crossed the river at right angles to enable logging on either side of the banks at the same time, this was deemed a higher priority for understanding the flows at the river.

This study was reliant on a complementary Sellafield project to obtain river stage information on the river Ehen within the same time frame. Unfortunately, even though some data were obtained from loggers installed along the river, the river surveying to allow river stage to be ascertained was not possible due to technical problems with the installed equipment. Therefore, calculation of head gradients between the river and the groundwater system cannot be determined accurately. As an alternative, there is an Environment Agency river gauging station 3km north of the site at Borough Hill near Braystones (ID:744130) and open source river level data from November 2012 onwards were downloaded. These data allowed approximated comparisons to be made with the logging data collected from the monitoring wells on either side of the river (BH747; BH747; BH6975) during 2012-

2013. It is recommended that this experiment is attempted again in future with river and monitoring well loggers recording simultaneously.



Borehole ID	Logging time period	Borehole depth (m)	Elevation (m)	Screen Interval (m)	Dip to Water (mgbl)	Depth of Logger (mgbl)	Logger Serial Number	Drift deposits (mbgl)	Sandstone (mbgl)	Location
BH745 p1	23/11/2012-11/07/2013	8.5	7.17	2.6-8.5	2.84	4.5	184613	0-8.5	n/a	(east) bank of r.Ehen
BH747 p1	26/07/2012-12/11/2013	6.5	5.94	0-6.2	2.82	4.5	184614	0-6.5	n/a	(west) bank of r.Ehen
BH6911 p1	17/11/2011-26/06/2012	65.0	7.32	63-64	7.0	10.0	184613	0-5.50	5.50-31	central sand spit
BH6912 p1	20/07/2011-25/06/2012	40.0	13.95	38-39	9.35	15.0	184883	0-31.5	31.5-40	central sand spit
BH6919 p1	28/06/2013-28/11/2014	15.9	10.47	11.5-12.5	6.81	8.0	184883	no info	no info	east side of river
BH6975 p1	20/07/2011-17/11/2011	41.2	9.652	35-36	4.87	11.0	185207	no info	no info	north of sandspit & pipeline
BH6975 p2	17/11/2011-06/11/2013	41.2	9.652	13-14	7.08	11.0	185207	no info	no info	north of sandspit & pipeline
BH6977 p1	20/07/2011-25/06/2012	20.1	7.502	16.5-17.5	3.94	10	184614	0-20.1	n/a	south end of sand spit
BH6978 p1	20/07/2011-17/11/2011	20.8	15.252	18.8-19.8	12	17	184613	no info	no info	central sand spit

Figure 3-3 Set up parameters and locations for the automatic data logger deployment in beach groundwater monitoring wells at the Sellafield beach and river Ehen interface. Normal Tidal Limit (NTL) is marked.

3.3 Results and Discussion

The results in this section are presented as individual studies that were carried out at the dynamic coastal interface. The sections are structured by the two receptor areas that were investigated firstly the beach spring discharges and discussion on the groundwater dynamics at the beach face and then secondly discuss the tritium discharges to the River Ehen and interpretation of potential controlling processes.

3.3.1 Spatial distribution of tritium in beach springs: 2009-2018 observations

3.3.1.1 Location variability of beach spring discharges

A preliminary inspection visit to the beach foreshore at a low tide in 2009 identified relatively low flow beach springs along the gravel front of the foreshore and were characterised by a lateral area or 'seepage' front that flowed into small pools or channels (refer to Figure 3-1). 'Spot' water quality samples have been collected along the Sellafield beach up to 3km north of the legacy nuclear site between 2009 and 2018. A total of 405 samples were collected over the study period. Beach springs were identified on the foreshore in all sample collection rounds between 2009 and 2018. The locations of the springs and river samples were mapped by GPS and are presented in Figure 3-4.

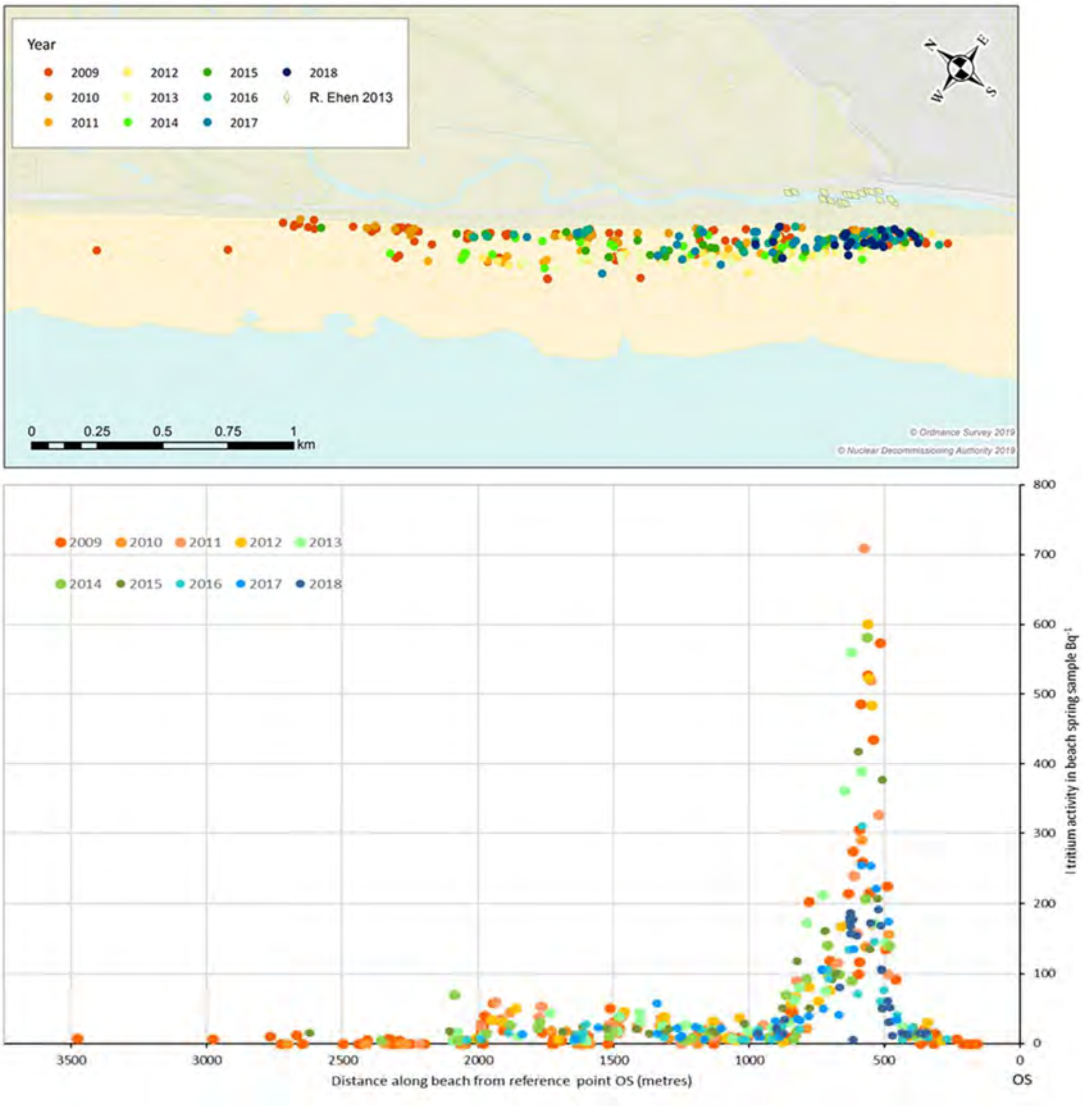


Figure 3-4 Annual beach spring sample locations from recorded GPS locations and tritium activity in samples as at each sample point 2009-2018. OS reference point coordinates 30.2267;50.2835

At the high-water mark line of the beach the loose gravel moved laterally and changed to steeper or gentler profiles across the beach during the study period depending on weather events, such as storms and strong tidal conditions. The springs appeared in relatively the same location despite the changing beach surface profile, with some minor lateral movement (up to 5-10 m) depending on the gravel front profile. Several distinct beach springs were seen at the Ehen spit on every field visit that appear to occur in clusters along the foreshore (Figure 3-4).

Figure 3-5 presents the annual variations in sample tritium activity found in spring waters surfacing on the beach for each sample round between 2009 and 2018. The tritium activity appears to reduce by approximately half over the 10 year period, which would be consistent with constant mass supply and a radioactive half-life of 12.32 years. The peak discharge concentrations at around 500 metres remains in a reasonably stable position throughout the 10 years of sampling. In 2018 limitation of resource (sampling personnel and laboratory analysis budgets) meant that samples were only collected from the spring peak discharge zone and therefore the data collected in 2018 is not reflective of the full width of plume footprint and should not be misinterpreted as a reduction in the width of the plume.

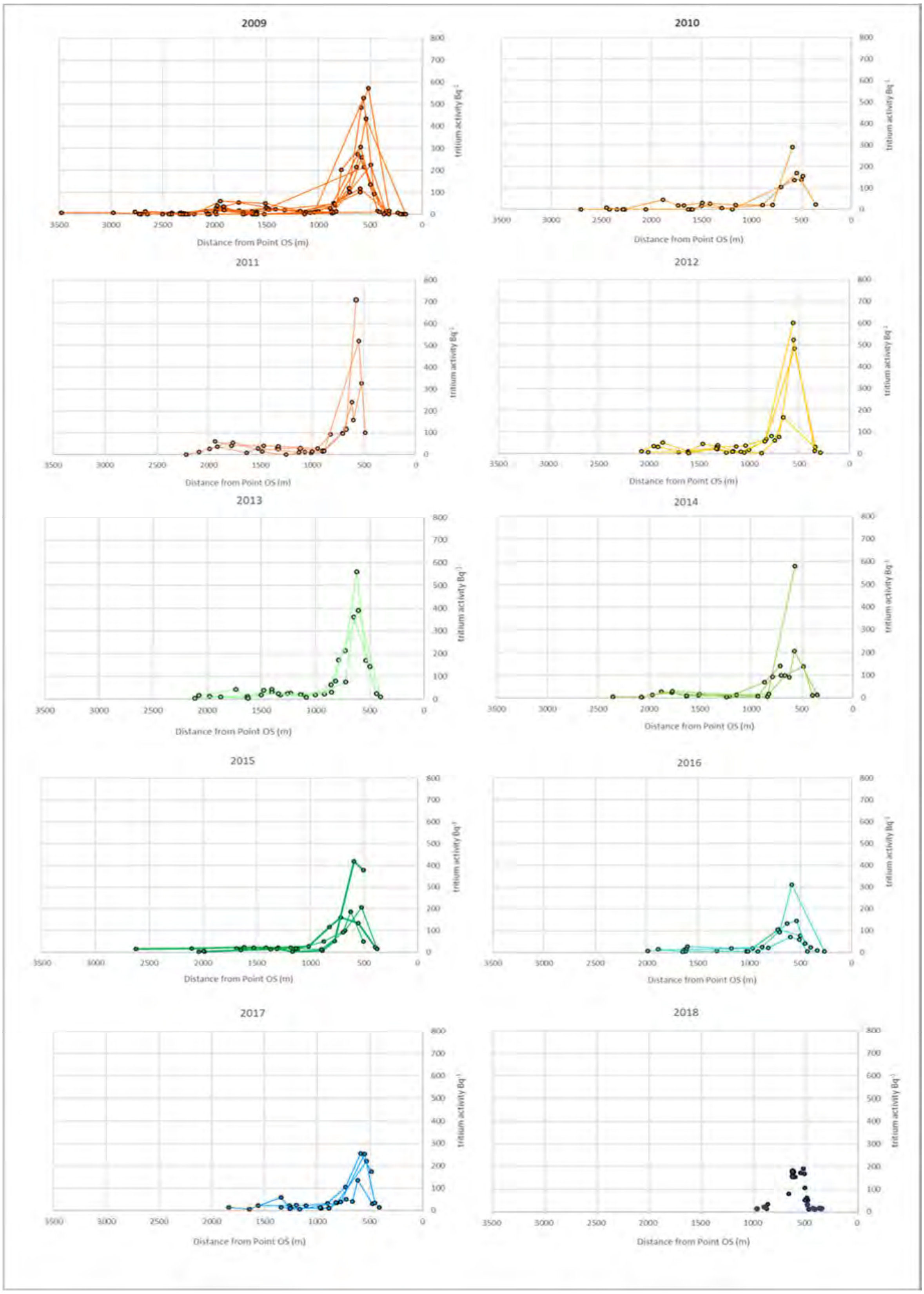


Figure 3-5 Annual lateral variations in beach spring locations and sample tritium activity Bql^{-1} for each sample round collected 2009-2018. Note that in 2018 only samples from around the higher concentration discharge point were taken.

3.3.2 Beach foreshore tritium discharge footprint

The tritium activity in the samples collected and analysed delineate a distinct asymmetric groundwater plume discharge footprint stretching 2600 metres northwards of the site along the foreshore (Figure 3-6). The lateral extent of the tritium groundwater plume was further north than expected based on the review of the historical data sets in Chapter 2, however after ten years of sampling it is concluded that the lower concentrations on the shoulder of the plume are real as they are consistently above background levels (5-10Bql^{-1} in local waters) This could have implications for future risks posed by more attenuated radionuclides, given the extent of plume expression northwards along the beach, the locations of the beach spring clusters within the footprint are used to further investigate groundwater flow pathways by particle back tracking in the Sellafield groundwater flow model in Chapter 5. The background seawater and surface water (river and local tarns) samples were analysed for tritium and did not have tritium activity above the limit of detection (minimum detectable activity).

The plume shape can indicate the type of groundwater flow regime transporting a contaminant to the receptor. A high velocity flow rate with low dispersivity potential will lead to a high aspect ratio (length to width ratio) of perhaps 10:1 or higher. A low velocity rate with high dispersivity potentially will yield a short broad plume with a low aspect ratio, the source width is also an important factor as well for plume width characteristics. In the case of this plume the maximum width at the beach springs is estimated at 2600m with a length from the source of approximately 800m, which provides an aspect ratio of about 3:1, the main source width is approximately 82 metres indicating relatively high dispersivity given the short distance between the source term and the beach. Controls on the asymmetric plume shape may be explained by firstly the amount of recharge or upgradient flow to the plume as it travels from the source. Alternatively, and perhaps more likely the footprint may be several thinner plumes with different source origin overlapping, with the main Windscale trenches leaching the highest tritium concentrations which creates the high sided peak directly down-flow of the trench location. There is also a buried channel under the site that may infer some preferential pathway (heterogenous glacial deposits), and the definition of this has been an ongoing concern and may be contributing to the pull of the flow, the river Calder boundary condition or possibly faulting may also influence the shape of the plume. Figure 3-6 shows a definite shoulder on the main peak, and then there are also several possible smaller peaks where there possibly appears to be high areas. This would fit with the multiple sources hypothesis and the smaller peaks would be likely as the travel distances from site significantly greater and possibly less direct.

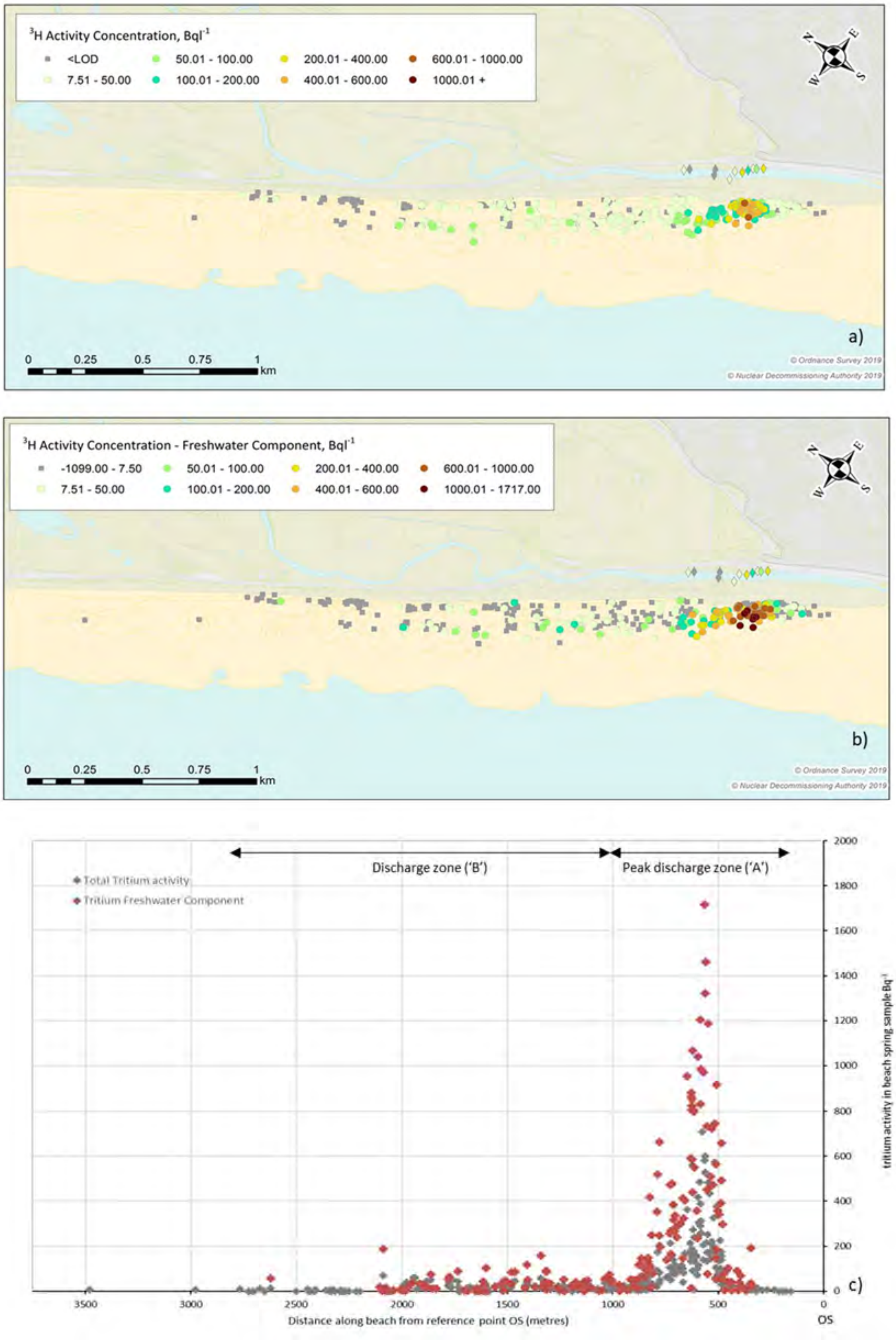


Figure 3-6 Tritium plume discharge footprint 2009-2018. A distinct asymmetric plume of 2500m width, with the spring peak discharge zone area 'A' recording the highest tritium activity levels, a) sample tritium activity found in the spring water samples; b) calculated tritium in spring water freshwater component; c) tritium activity versus distance along the foreshore from basepoint 'OS' (30.2267,50.2835)

3.3.3 Beach springs – variations in temperature, pH and specific conductance

Field measurements for temperature, pH and specific conductance (SPC) were recorded for all the tritium beach spring sample locations between 2009-2018 and are plotted against distance along the beach (from point OS) in Figure 3-7. Temperature of the samples ranged from 1°C-23°C (average 11.8 °C), with colder temperatures occurring during the winter months, and the highest temperatures in the summer months, and were similar temperatures across the plume width at each sample round, i.e., no distinct peaks or changes in temperatures against distance. pH readings remained stable throughout the sampling period with most of the samples in the range of 7.5 to 8.4, typical of brackish waters (Figure 3-7).

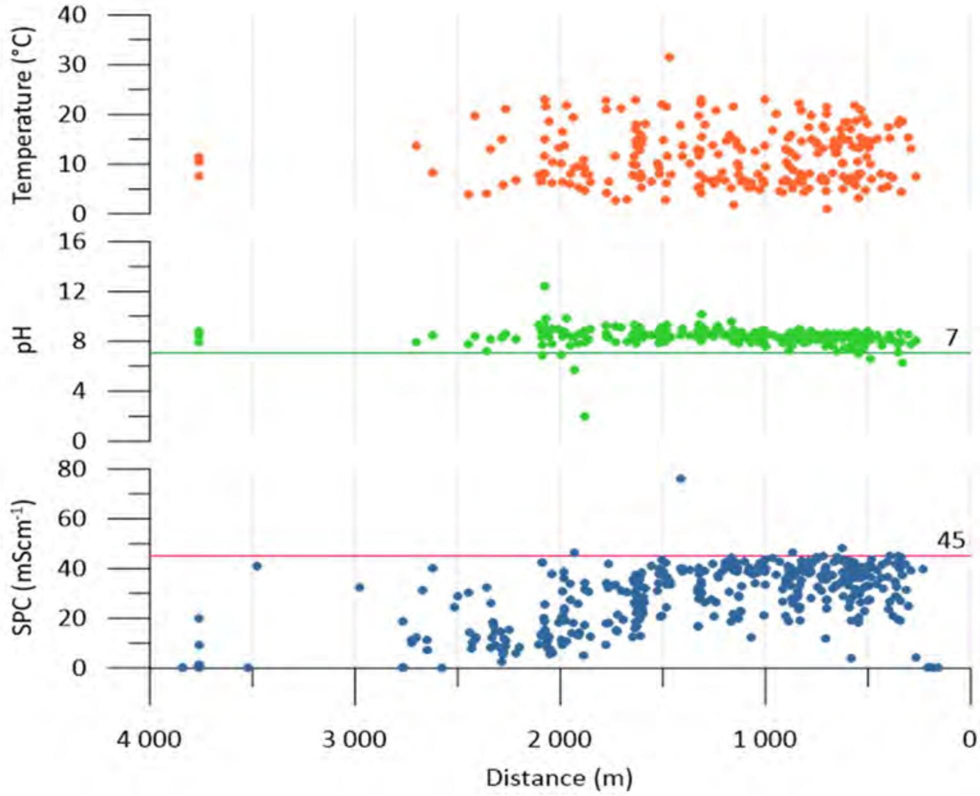


Figure 3-7 Field measurements of SPC, pH and temperature of the beach springs sample locations plotted against distance from point OS.

SPC readings varied over the sampling period and decrease northwards. This may suggest less saline waters in the beach spring flows, and help explain the asymmetric nature of the plume, as the lower

SPC readings correlate with the plumes lower tailing edge of tritium activity concentrations (Figure 3-8). No distinct relationship is seen between the pH and temperature against tritium activity concentrations and are interpreted as seasonal ambient fluctuations.

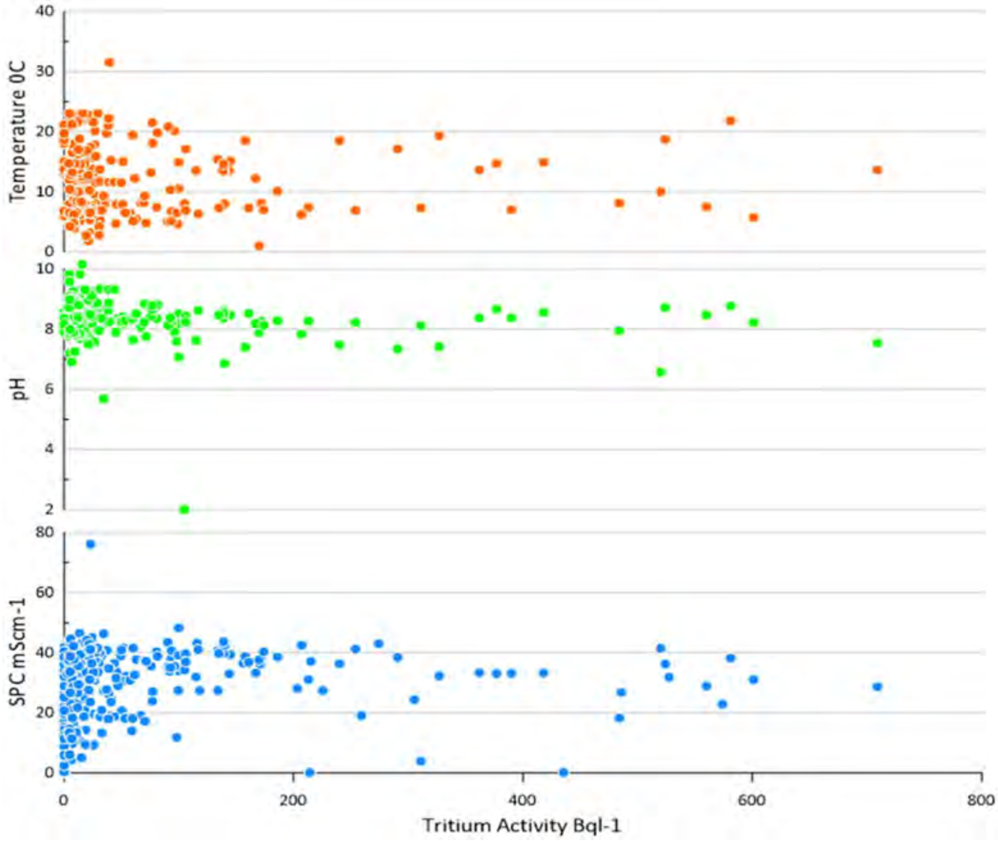


Figure 3-8 Field measurements SPC, pH and temperature of the beach spring samples plotted against sample tritium activity

3.3.4 Temporal variations of tritium activity in beach springs

3.3.4.1 Time-series of tritium activity in beach spring (confluence) 2009-2018

Descriptive statistics for the tritium activity samples collected between 2009 and 2018 are summarised in Table 3-1. Tritium beach spring sample results ranged from undetectable (below limit of detection of 7.5 Bql^{-1}) to a maximum of 709 Bql^{-1} (as found at the beach surface) and 1717 Bql^{-1} (in freshwater component) across the plume discharge footprint. All results were considerably below the WHO drinking water standards of 10,000 Bql^{-1} and considered very low risk to the public.

Table 3-1: Descriptive statistics of tritium activity (Bql^{-1}) in beach spring area on Sellafield Beach from 2009 to 2018. Results below <7.5Bql^{-1} were below the analytical limit of detection and marked <LOD in table.

		Tritium activity Bql^{-1}				Tritium as freshwater component Bql^{-1}			
Year	Samples	Max	Min	Mean	St-Dev	Max	Min	Mean	St-Dev
2009	115	573.9	<LOD	48.9	108.4	1589.83	<LOD	632.90	832.93
2010	29	290.8	<LOD	43.4	70.6	1285.27	<LOD	881.57	337.52
2011	36	709.0	<LOD	83.9	149.1	1269.41	<LOD	1102.63	280.38
2012	37	601.0	<LOD	71.9	144.0	1462.2	<LOD	160.1	362.9
2013	34	560.0	<LOD	80.2	127.0	1205.0	<LOD	187.0	316.1
2014	30	581.0	<LOD	61.2	111.0	1717.2	<LOD	153.4	441.6
2015	34	418.0	<LOD	67.5	100.0	1039.6	<LOD	180.0	279.3
2016	30	311.0	<LOD	46.4	65.1	984.7	<LOD	131.4	230.6
2017	31	254.0	<LOD	55.4	73.0	832.2	<LOD	144.0	241.6
2018	29	191.0	<LOD	91.0	74.1	881.2	<LOD	379.4	354.0

The time-series data for tritium activity collected at the prominent beach spring (peak discharge zone), the peak of the tritium plume are presented in Figure 3-9 for tritium activity and the calculated tritium activity in the freshwater component of the beach spring (based on the percentage seawater) over nearly a decade (2009-2018). Tritium levels appear to be reasonably stable at lower concentrations, fluctuations possibly due to seasonal or tidal response. There are fluctuations in concentrations over the time period with an oscillation of approximately 1000 Bql^{-1} in magnitude, which may be seasonal or related to rainfall recharge.

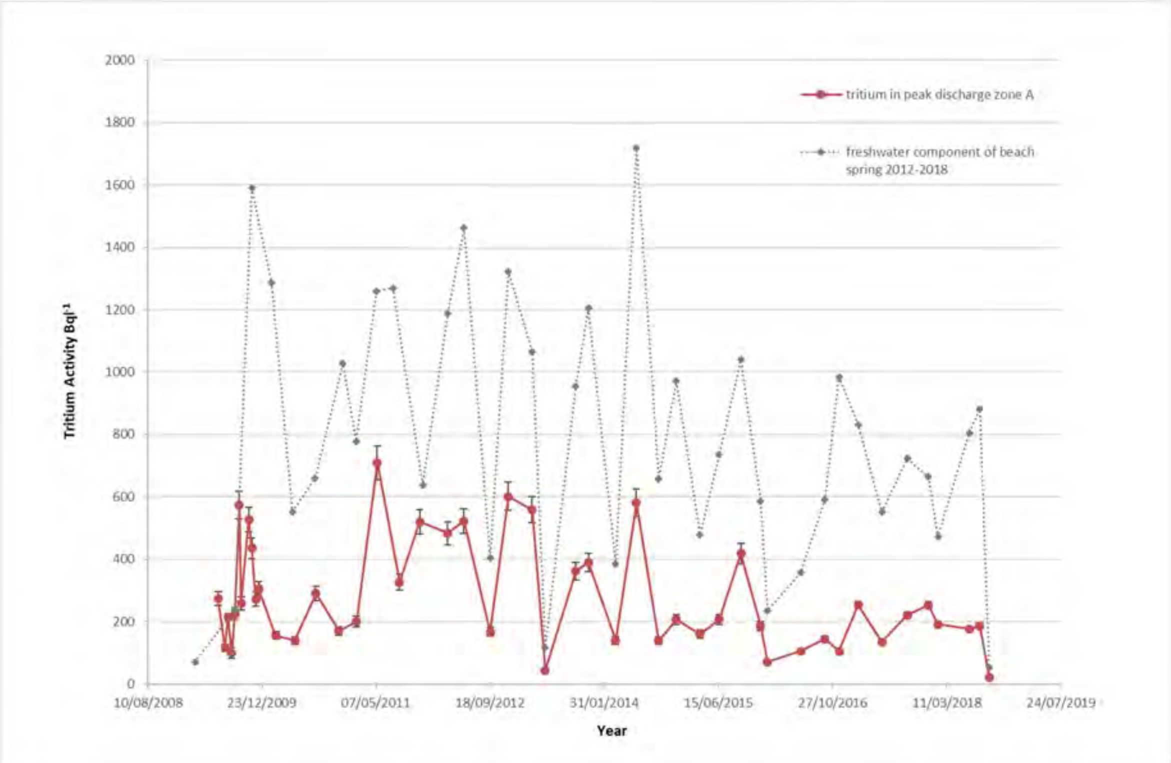


Figure 3-9 Time-series of tritium activity in samples in peak discharge zone 'A' collected between 2009-2018

3.3.4.2 Long term temporal profile of tritium activity in peak discharge area 'A' (1975-2018)

The time series graph for tritium from 1975 to 2018 has been constructed from data found in historical reports (presented in Chapter 2) combined with the new field data collected that took place from 2009 to 2018 from the peak discharge zone ('A') beach spring area (Figure 3-10). The primary tritium plume from the Windscale trenches has travelled towards the beach from 1952, Figure 3-10 indicates that it broke through at the beach spring receptor prior to 1975, as the time-series graph shows a steep downward trend suggesting the peak of the tritium pulse has already passed by the time the first beach and borehole samples were collected in 1975. A steep decline in tritium activity has occurred from 1976 and 1990, with a number of smaller peak events in 1984, 1986, 2003, and between 2009 and 2011, which may be attributed to other source term signatures appearing at the beach, but relatively low in magnitude compared with the dominant Windscale trenches low tailing concentrations.

Another explanation is as the PVC wrappings around some of the waste in the trenches deteriorates, intermittent higher concentration pulses may enter the groundwater.

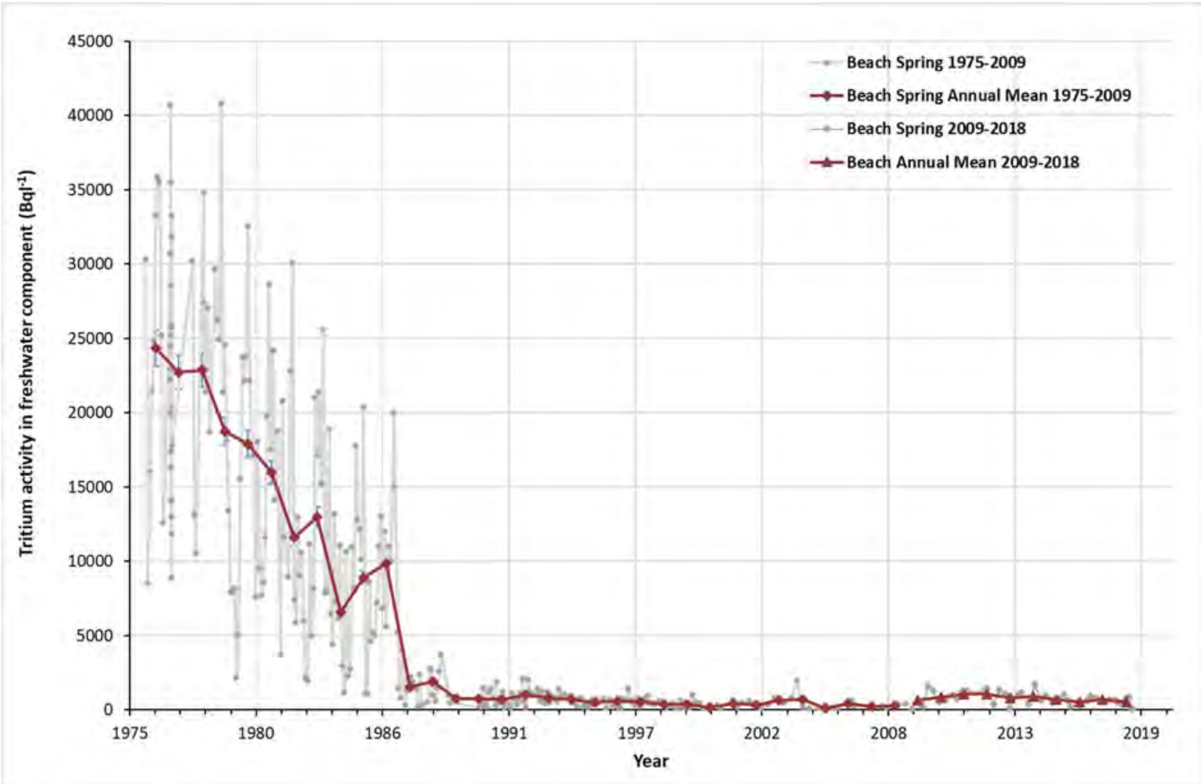


Figure 3-10 Time-series of tritium activity as freshwater component in the peak discharge zone 'A' 1975-2018.

The overall trend also correlates reasonably well to a radionuclide half-life of 12.32 years, over four decades (roughly half the activity magnitude every 10 years), and shows some decline after the initial high peak and the data may indicate that it has been a fairly constant flux of tritium to the beach springs after the waste was emplaced between 1952-1964 when the tritium concentrations are normalised to account for radioactive decay to the last sample date in 2018 and indicates that although the plume looks stabilised it is not yet in decline (Figure 3-11). Alternatively, if we consider the multiple plume hypothesis the constant source trend may be the sum of the plumes.

The new data from 2009-2018 supports the original conclusions made in Chapter 2 that the tritium plume(s) have stabilised. A further review inland from the beach to the site of the tritium activity in the groundwater monitoring wells was carried out to identify if there is any further risk of other larger tritium pulses migrating towards the beach (discussed in Chapter 5) and aim to monitor their arrival at the beach springs to support transport travel time understanding.

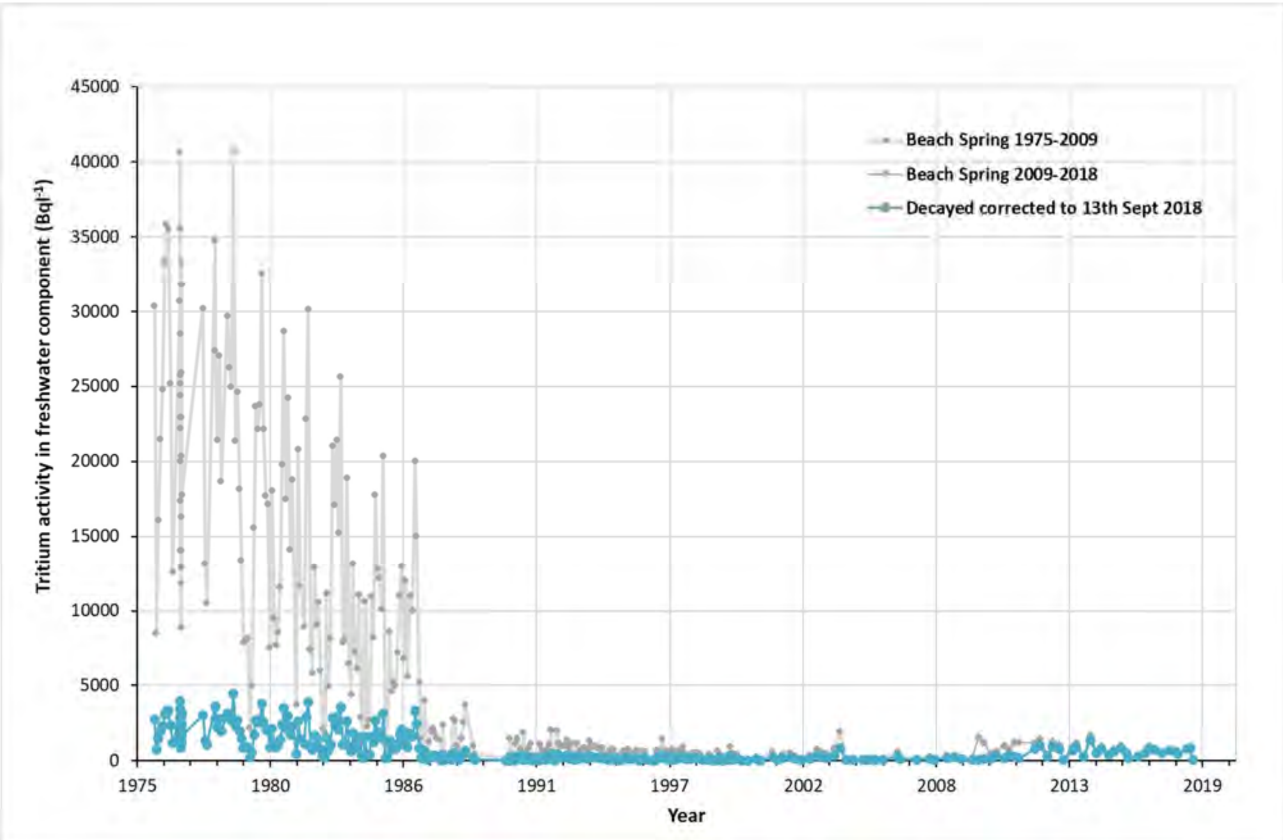


Figure 3-11 Tritium data from peak discharge zone 'A' 1975-2018 half-life decay corrected to the most recent sample date of the 13th September 2018.

3.3.4.3 Seasonal fluctuations in tritium activity (2009-2018)

There are no obvious spatial seasonal fluctuations in terms of the tritium in samples (Figure 3-12) or freshwater tritium (Figure 3-13). Spring and autumn are often drier months in this region of the UK and show slightly more elevated tritium activity results for both sample and calculated freshwater component and may suggest therefore that lower tritium activities occur during seasons of higher precipitation and affected by dilution.

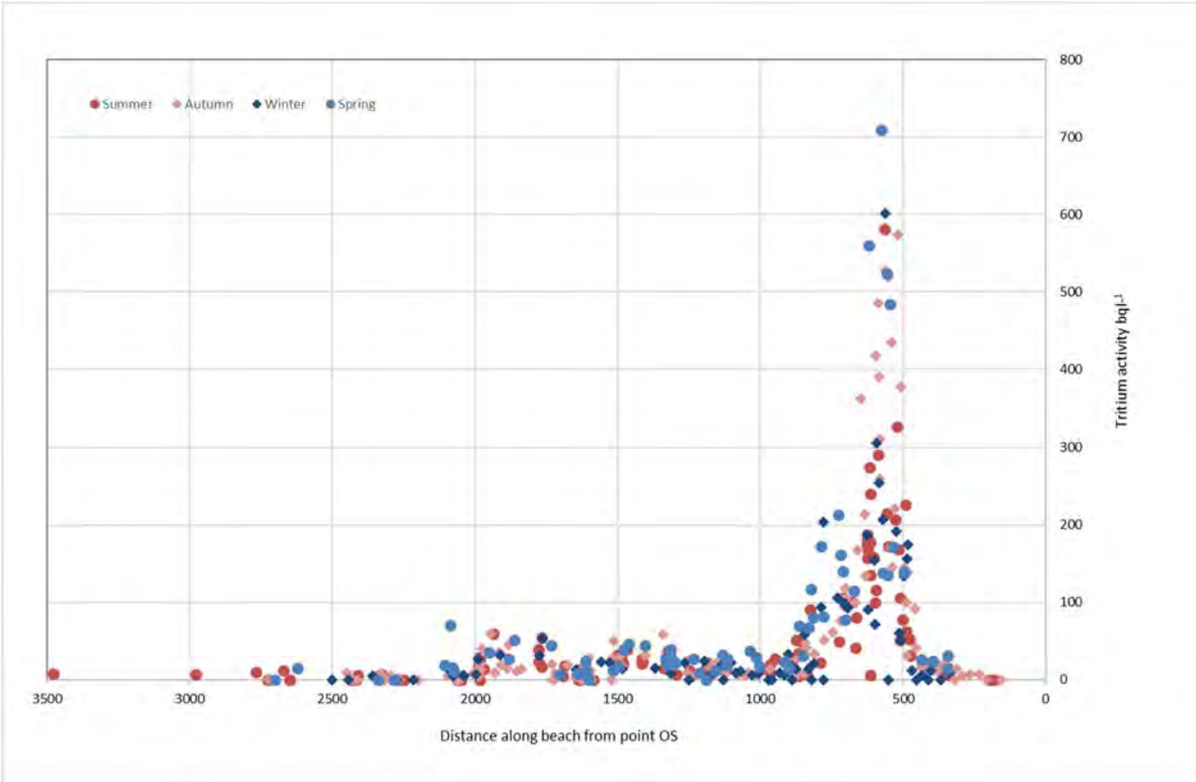


Figure 3-12 Seasonal fluctuations in beach spring sample tritium activity 2009-2018. Sample tritium activity is plotted in the seasons as Spring (Mar-May); Summer (Jun-Aug); Autumn (Sept-Nov); Winter (Dec-Feb).

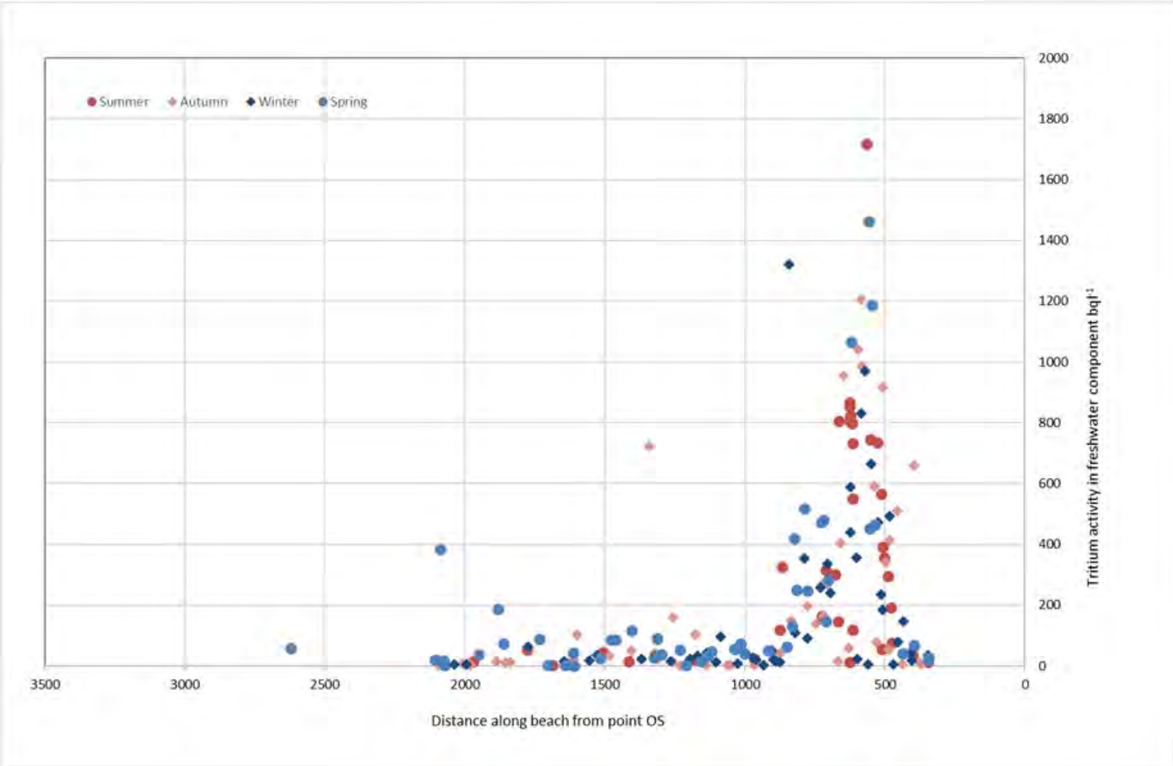


Figure 3-13 Seasonal fluctuations in tritium activity as freshwater component of beach spring water samples 2009-2018. Tritium activity is plotted in the seasons as Spring (Mar-May); Summer (Jun-Aug); Autumn (Sept-Nov); Winter (Dec-Feb).

These observations can be further explored by considering the local rainfall data and the associated SPC measurements to understand whether the beach springs contain a higher proportion of freshwater, and thus a lower SPC value. Figure 3-14 plots the mean monthly rainfall and beach spring SPC field measurements. There appears to be a slight lag effect but in general after higher periods of rainfall the SPC reduces to be less brackish in the range of 20 to 30 $mScm^{-1}$ and the SPC rises during drier periods towards 35-45 $mScm^{-1}$. The observations do not provide a convincing strong trend, and possibly higher resolution data would be able to draw better conclusions.

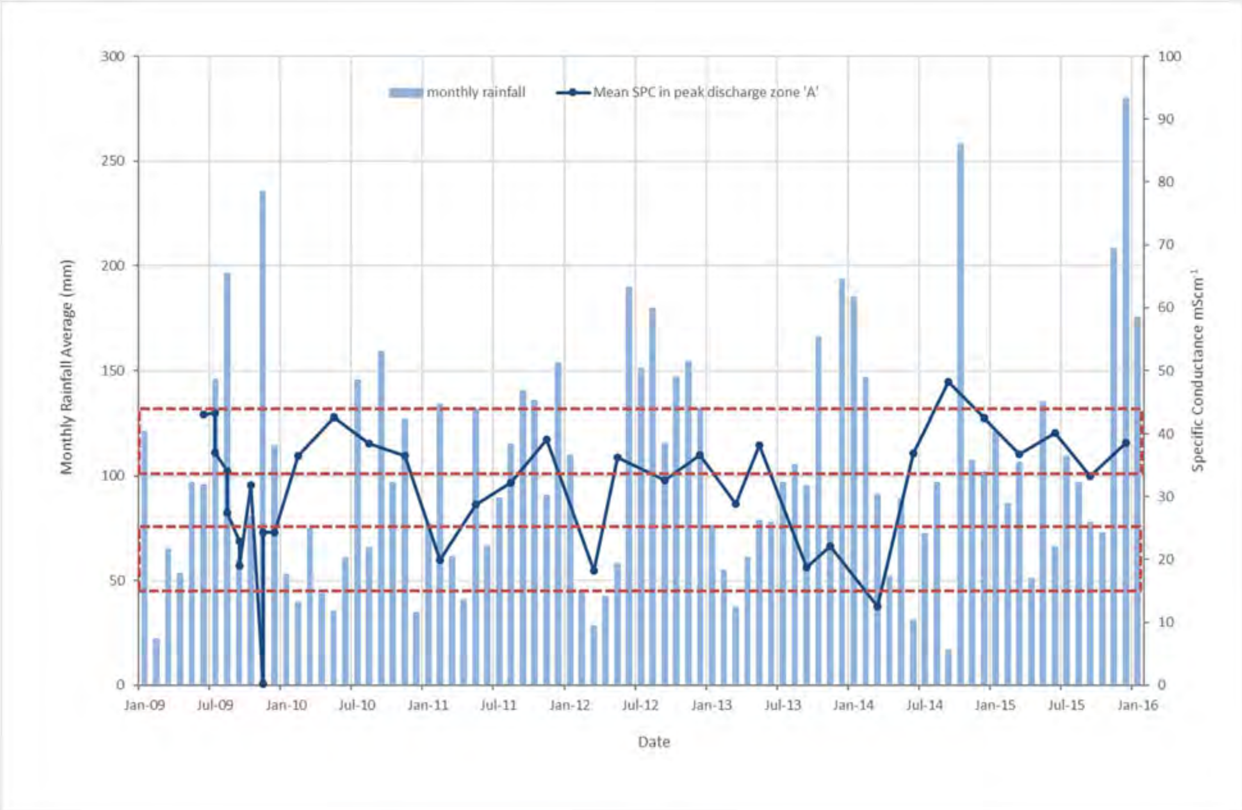


Figure 3-14 Average monthly rainfall in the study area versus the SPC measurements taken at the beach spring peak discharge zone 'A'.

3.3.4.4 Tritium activity in beach peak discharge area 'A' and response to rainfall

Tritium activity versus average monthly rainfall is shown in Figure 3-15. The time-series from 2009 to 2016 was analysed so that comparisons could be drawn with the rainfall versus tritium activity profile presented in the previous historical Section 2.6 for 1980-1986. A convincing trend between the tritium and the rainfall is hard to determine in Figure 3-15 and tritium concentrations may look to increase slightly in the springs during drier periods, with some lag effect. The calculated tritium in freshwater component of this peak discharge zone spring is also presented and has similar fluctuations. The temporal data seen in the archive data (Figure 2-11) was at higher temporal resolution and it may be the case here that the temporal resolution is not high enough to see the controls, and likely that both rainfall and tides are both important and hard to tease apart in this coastal environment. There are a number of issues that could cause the variability in concentrations other than just rainfall/recharge, such as tidal cycles, the exact sampling time (relative to the results present in Figure 3-15), spatial lateral movement in plumes, discharge flow field variations and the peak discharge zone samples varying in proportions sampled and therefore this complexity means there are no firm conclusions drawn on the response of tritium to rainfall.

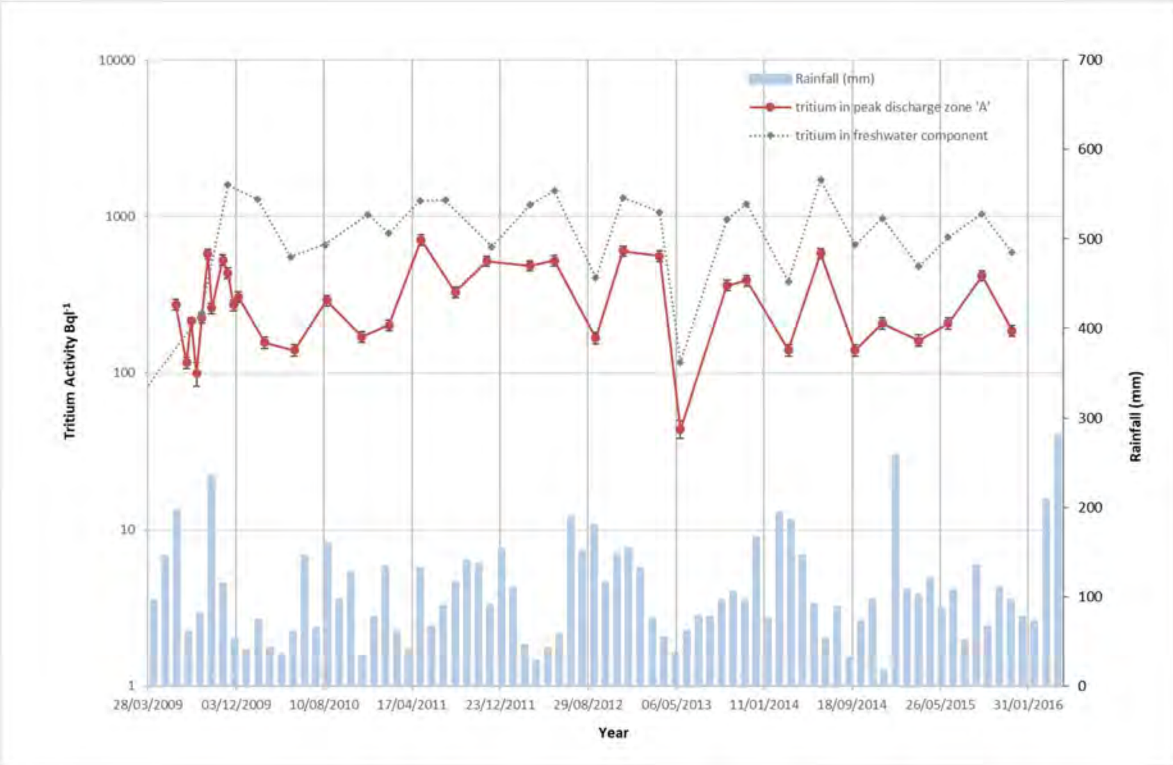


Figure 3-15 Response of tritium to average monthly rainfall fluctuations 2009-2015.

3.3.5 Tidal response of beach spring discharges

3.3.5.1 Beach spring discharge and head characteristics

Discharge rates, head and electrical conductivity (EC) was measured at six beach springs between the 21st July and 1st August 2017 by Hibbert (2017). To investigate the discharge rates of the springs two methods were used: the V-notch weir method and the velocity area (float) method. The results are presented in Figure 3-16. The average discharge rate for the V-notch weir method was 0.56 ls^{-1} (range of 0.022-1.25 ls^{-1}) and a higher rate of 3.88 ls^{-1} for the float method (range of 1.3-11.3 ls^{-1}), showing a weak correlation between the two methods, even when correction for the velocity is lower than the surface velocity.

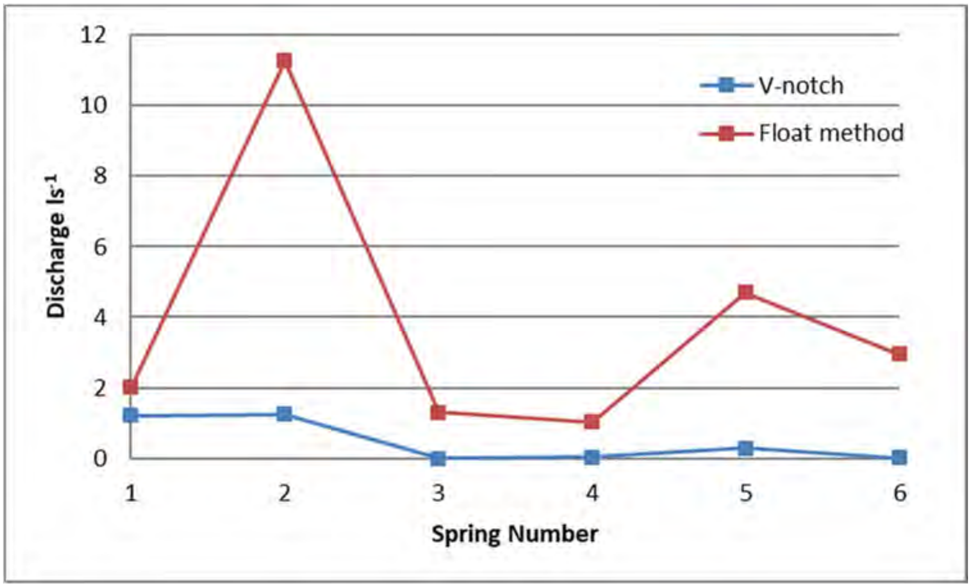


Figure 3-16 Comparison of average discharge results from the V-notch weir and float methods at the six beach spring locations along the Sellafield beach (from Hibbert 2017).

The range in results may have been attributed to both methods requiring significant excavation to build both the V notch weir and a straightened 5 m channel and as such may have been collecting a wider and different discharge baseflow. Timing a piece of seaweed floating in the channel may have led to some inaccuracies: a neutrally buoyant float of known size and surface area might have given more consistent results. An alternative '1 litre jug' method is used in the later experiment on 8th August due to the low flow of the seepages to provide an alternative representative flow rate at the beach face (Section 3.3.5.2).

Figure 3-17 shows the time-series of falling head for the peak discharge zone 'A' in response to the falling tide on the 1st August 2017 (Hibbert, 2017). Interestingly, the head continues to fall whilst the tidal direction reverses and starts to rise again until the point the beach spring is submerged by the incoming tide. Hibbert (2017) reports a strong correlation of flow and EC where falling heads initially suggest saltwater drainage. However the conclusions made around the beach springs having a high freshwater percentage (up to 99 %) are not convincing as this is not consistent with the reported spot EC measurements in an average range of 31.7-38.5 $mScm^{-1}$ which tends to indicate fairly brackish waters (typical ranges of freshwater are 0-1.5 $mScm^{-1}$ to seawater at 45-60 $mScm^{-1}$ given in Rusydi,2018). A further limitation of the study is that monitoring EC in the field is sensitive to temperature (about 2.5% per degree), and given the measurements were taken on different days, times and locations Specific Conductance normalised to 20°C would be more accurate for data comparisons.

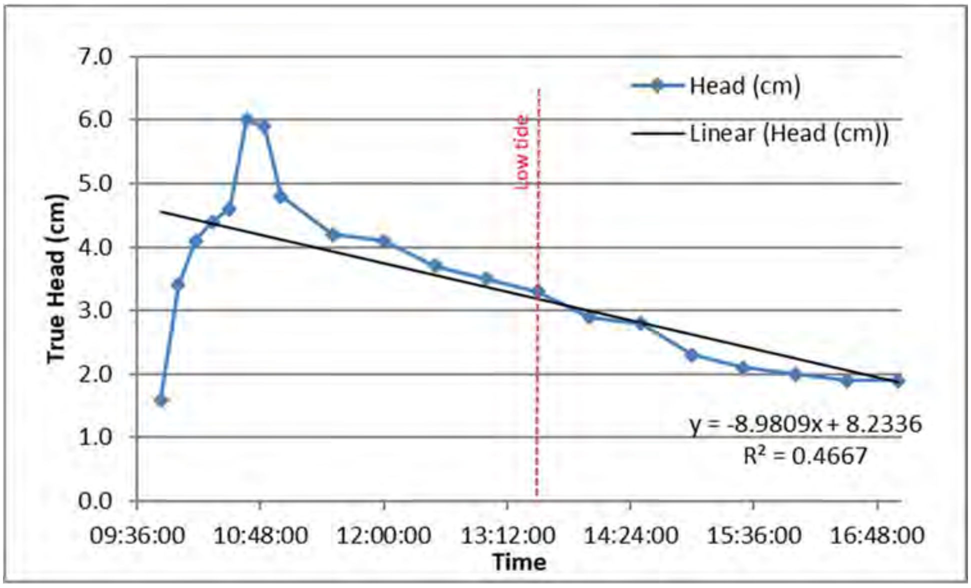


Figure 3-17 The measurement of beach spring 1/peak discharge zone (54.4146248, -3.5112948) head in response to the falling tide on the 1st August 2017. High tide recorded at 06:57am and to low tide at 13:34pm (from Hibbert, 2017)

During the 5 day observation of the beach springs, Hibbert (2017) attributes a combined decrease in salinity in the spring waters at equivalent tidal state to perhaps indicate a diminishing presence of a saline cell and is supported by a reduction in spring flows over the five days during the neap cycle (Abarca et al., 2013) and broadly is aligned with the archive and field observations. A more detailed investigation would be required to understand these dynamics and therefore it would be recommended to install a series of shallow beach monitoring wells with automatic logging and a geophysics survey.

3.3.5.2 Beach spring tritium activity fluctuations in response to tidal influence

An experiment was carried out on the 8th August 2018 to replicate the 1977 experiment discussed in Section 2.5.2. The purpose was to study the short-term variations in tritium concentration and salinity in the spring peak discharge zone 'A'. The tritium activity was monitored at regular intervals following low tide until the beach spring became submerged by the rising tide. The relationship between the tritium activity and the tidal fluctuations is given in Figure 3-18. The tritium activity increased over the period, whilst the percentage seawater in the beach spring sample decreased. A similar trend of a rising tritium activity in the freshwater component of the sample was also seen, which would indicate more groundwater flow was surfacing at the beach as the effects of the saline cell discharges reduced.

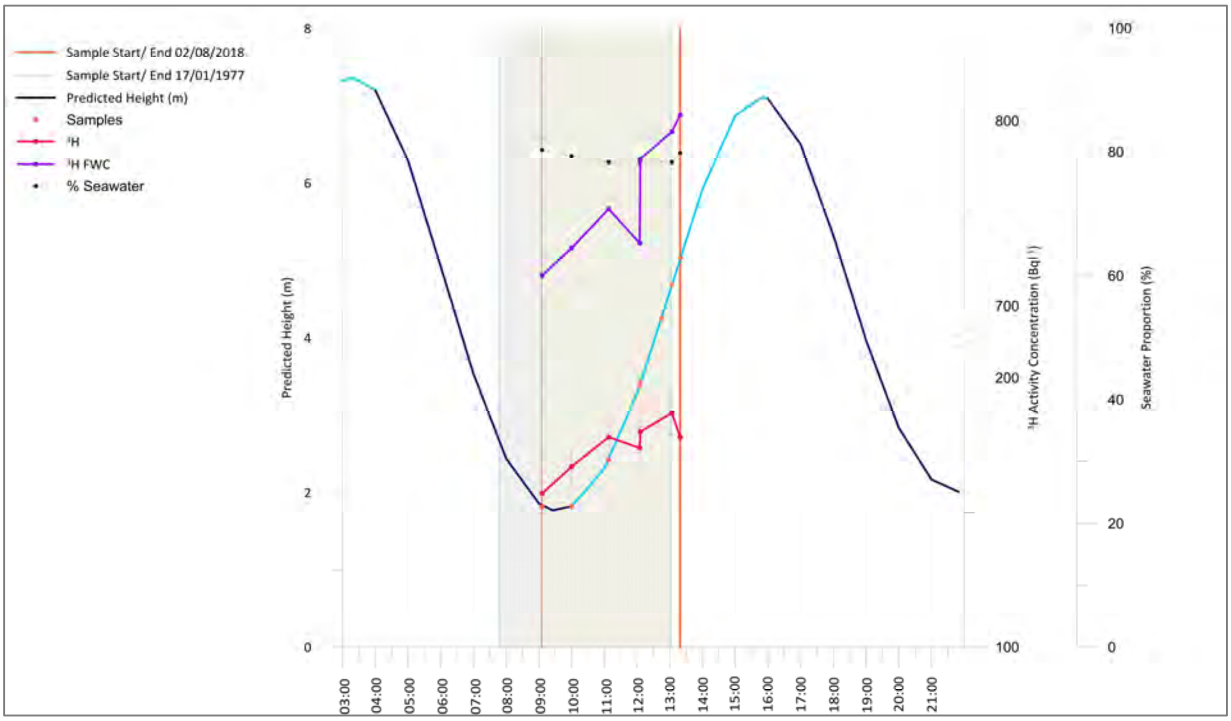


Figure 3-18 Results of the sample tritium activity collected from low tide (09:30am) until the beach spring became submerged by the tide (13:22pm). Predicted height is the estimated tidal elevation.

The comparative tidal cycle sampling window for 1977 is overlain and the results have been converted to the equivalent Bql^{-1} readings for tritium activity in Figure 3-19. This earlier experiment also showed a response of increased tritium activity accompanying a decline in the percentage of seawater. Interestingly the tritium activity in the freshwater component shows a declining trend until the point when the beach spring becoming submerged. This is harder to explain as the drop in seawater content would suggest a greater proportion of groundwater flow. Calculations of the freshwater component may have differed between 1977 and 2018 and this may also affect the trends seen.

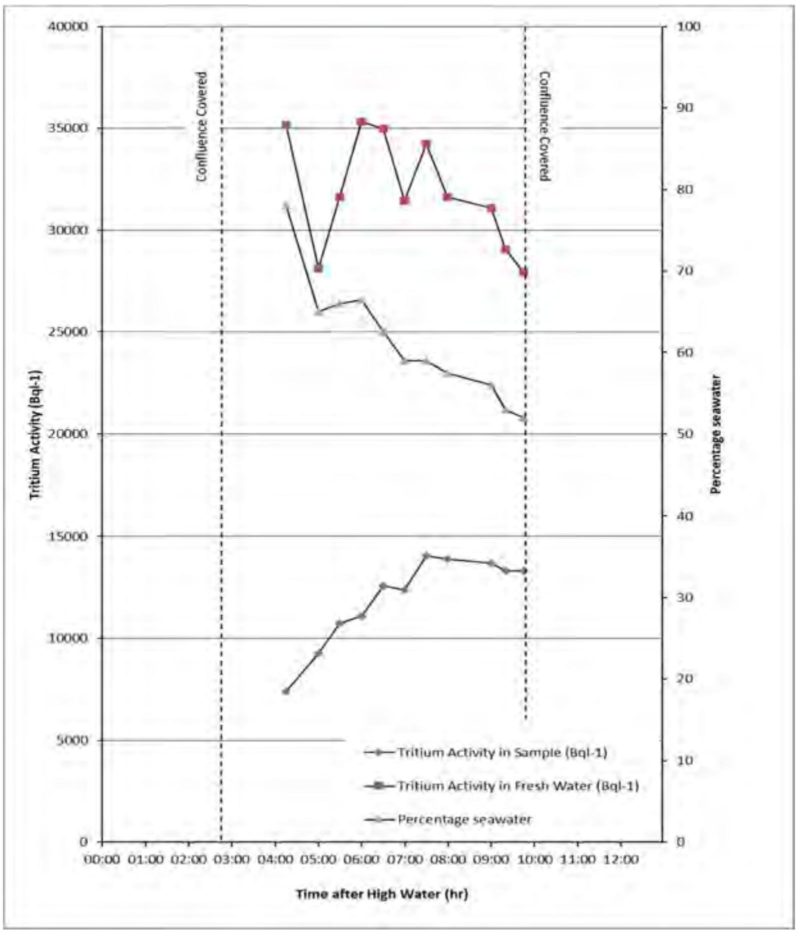


Figure 3-19 Tritium activity during falling tide reproduced and converted to Bql^{-1} from Whittaker (1977) from the first experiment carried out in 1977 as a comparison to results taken in 2018. The dashed lines of the sampling window are overlain in the previous Figure (3-18)

3.3.5.3 Beach spring tritium discharge rate

Beach spring discharge flow rates recorded on 8th August 2018 are presented in Table 3-2. Flow rates for the beach spring by the '1 litre jug' method produced an average of 0.0135 ls^{-1} which is comparable with the V-notch method results described in section 3.3.5.1. The tritium flux rate estimated was estimated as 5.18 $x10^{8}$ Bql^{-1} in 1976 and 2.89 x 10^{10} Bql^{-1} in 1977 by constant rate injection tracer method. The average tritium flow rate was estimated from the 2018 results at 2.04x 10^{05} ld^{-1}, which is significantly reduced from the measurements made in the late 1970s even when taking in to account the radioactive half-life of tritium. There is a slight drop in the SPC values, as expected, with the slight drop in percentage in seawater shown in Figure 3-18. pH remains relatively stable around 8.5 to 9.0. The temperature drops to its lowest at low tide and then rises by over 1.0 °C as the tide rises.

Table 3-2 Field measurements and calculated discharge results from beach spring flow measurements and tritium activity analysis between tides on the 8th August 2018, Low water recorded at approximately 09:30am and beach spring became submerged by seawater at 13:22h.

Sample Time	Estimated Tidal height (m)	SPC ($mScm^{-1}$)	pH	Temp °C	Ls^{-1}	La^{-1}	^3H Bq d^{-1}	^3H Bq a^{-1}
09:06	1.86	42	8.57	18.2	0.0079	2.50E+05	1.08E+05	3.93E+07
09:47	1.77 (LW)	41.7	8.75	17.7	0.0179	5.66E+05	2.59E+05	9.45E+07
10.38	1.82	41.46	8.82	17.8	0.0149	4.69E+05	2.29E+05	8.34E+07
11:40	2.42	41.4	8.82	18.5	0.0127	4.02E+05	1.98E+05	7.23E+07
12:40	4.25	41.3	9.06	19.6	0.0154	4.86E+05	2.49E+05	9.09E+07
13:15	5.03	41.17	9.13	19.8	0.0118	3.73E+05	1.82E+05	6.63E+07
Mean		**41.5**	**8.85**	**18.6**	**0.0135**	**4.24E+05**	**2.04E+05**	**7.45E+07**

Producing an accurate discharge rate is challenging given the low flow seepages, installation of seepage metres along a defined spaced discharge front would be recommended to produce a more representative discharge rate, with regular water quality samples to analyse for tritium activity. The results do however provide a crude estimate of the discharge rates from the prominent peak discharge zone beach spring, where the highest tritium activities are recorded. They could be used to improve the estimated flux discharges in the Sellafield groundwater flow model.

3.3.6 Temporal variation of tritium in beach groundwater monitoring wells

Figure 3-20 shows the time-series for tritium activity in beach groundwater monitoring wells to 2018, for both the east (Sellafield side) and west side (beach) of the river Ehen. Several of the monitoring wells installed in the late 1970s are no longer active or regularly sampled. The more modern '6900' series of monitoring wells located on either side of the river Ehen have been added to the time-series data set. The tritium activity concentrations have declined over forty years, with minor peaks that correlate well with the peaks seen on the beach springs temporal profile. Tritium activities consistently remain relatively low on the east side of the river compared with those seen along the west bank, thus providing further confirmation that tritium is surfacing along this key dynamic boundary between the groundwater, river and the beach springs.

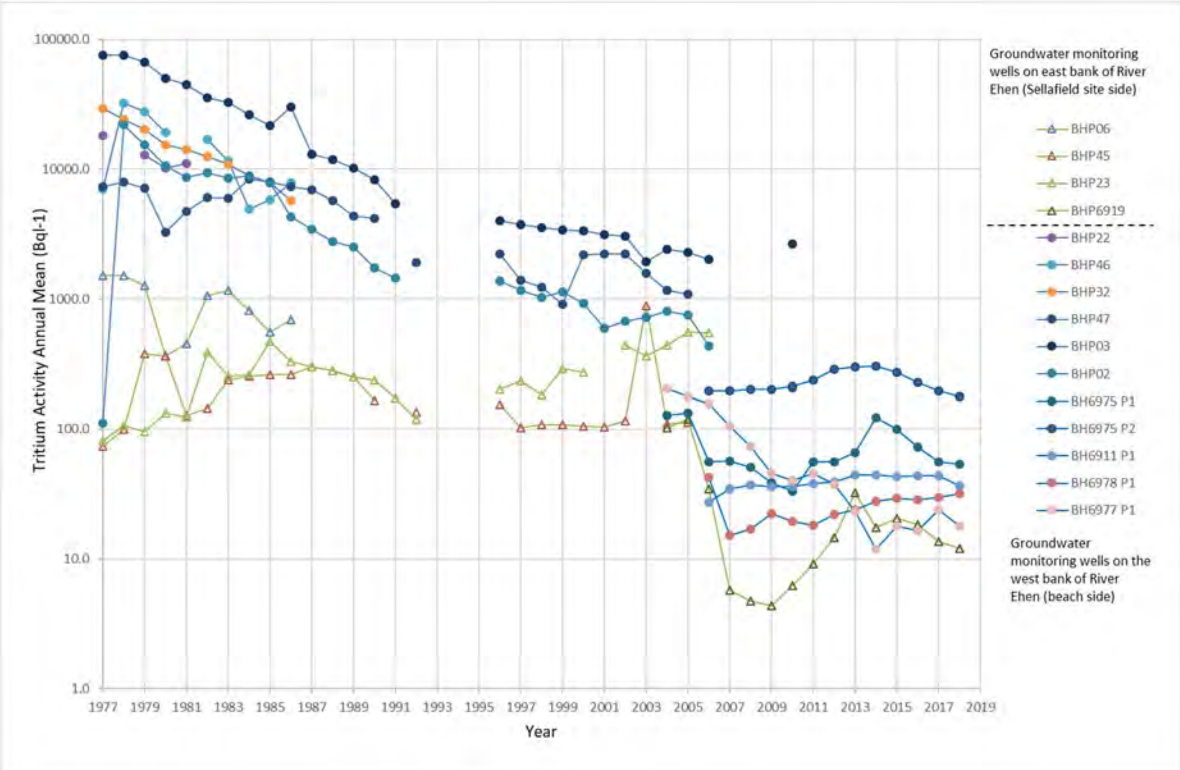


Figure 3-20 Time series profile of tritium activity (log-scale) of coastal groundwater monitoring wells located either side of the River Ehen.

3.3.7 Interpretation of beach spring groundwater dynamics

The groundwater in the beach area can be considered to an unconfined aquifer, dynamic and water flows through saturated and unsaturated sediments by tides, waves and swash, and other minor interactions with evaporation and deeper aquifers (Horn, 2002). Tritium acts as a perfect tracer in groundwater, and this study has confirmed that tritium has been surfacing in beach springs on the shoreface for over 45 years. This supports the comprehensive research concepts in beach groundwater systems summarised by Horn (2002, 2006). These concepts are adapted and represented in Figure 3-21 and defined below.

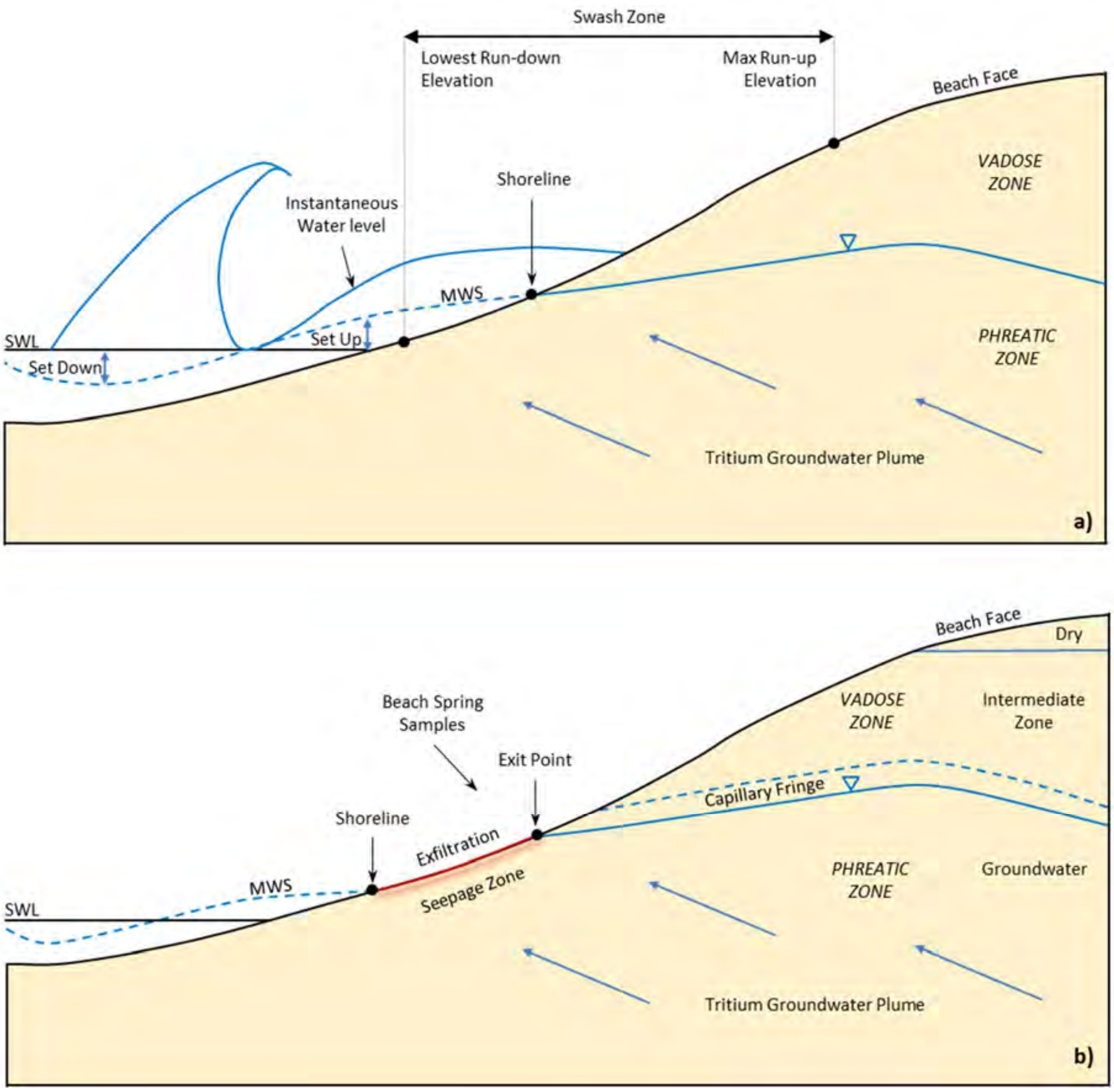


Figure 3-21 Sketch of beach spring groundwater dynamics (adapted from Horn, 2002;2006). a) Tide in: surface and subsurface water levels in the swash zone; b) Tide retreating: beach groundwater zones when the water table is decoupled from the tide.

Figure 3-21 shows the key phases between the complex interaction of surface and groundwater in the swash zone (a) and decoupling between the tide and the beach water table occurs when the groundwater exit point becomes separated from the shoreline (b) this occurs because the rate at which the beach drains is less than the rate at which the tide falls, so the tidal elevation generally drops more rapidly than the water table elevation and decoupling occurs (Horn, 2012). Below the exit point a seepage face develops where the water table coincides with beach face and is recognised by the glassy surface seen and sampled on the field visits to the beach near Sellafield. The observations of tritium in the seepage samples presented in Section 3.3 supports this dynamic beach groundwater model.

Figure 3-21 shows where the mean water surface (MWS) intersects the shoreline and the still water line (SWL) which represents a still water level in the situation of no waves. Changes in water levels (and hydrostatic pressure) are known as set up or set down. The phreatic zone is the fully saturated zone below the water table, while the vadose zone is the unsaturated zone between the water table and the beach face. Pore waters are negative in the capillary fringe area on Figure 3-21(b) and Horn (2012,2016) concludes that beach groundwater zone should be considered instead by water pressure distributions (rather than saturation).

3.3.8 Spatial distribution of tritium in the River Ehen

Figure 3-22 shows the results of the 'spot' samples taken in 2013 along either side of the riverbank. Samples from the centre of the riverbed were hard to attain due to the highly consolidated gravel, which could not be penetrated with a trowel or hand auger. The collected samples were separated for liquid and sediment at the laboratory and analysed for tritium activity. The results shown in Figure 3-7 need to be interpreted with a little caution as a regular grid sampling was not possible, and therefore the spatial distribution of the samples was determined by the access (depth of water) and collected from areas where the sediment was less consolidated. The maximum recorded tritium activity in the samples was 4970.35 Bql^{-1} in the liquid component and 1108.08 $BqKg^{-1}$ in the sediment component of a sample on the east bank (Sellafield side) approximately 100m south of the pipeline.

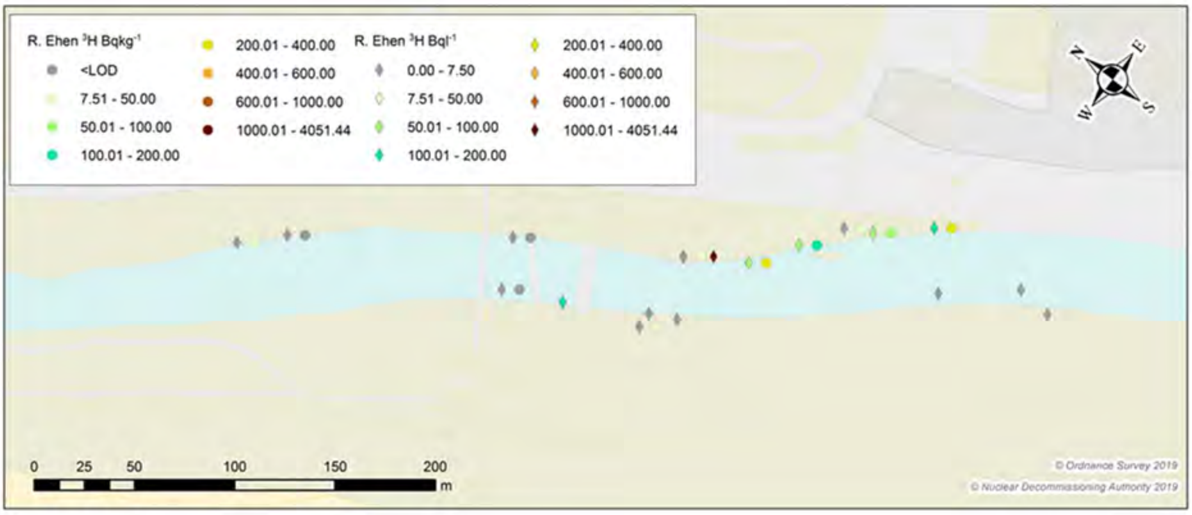


Figure 3-22 Spatial distribution of tritium activity in river sediment ($BqKg^{-1}$) and liquid samples (Bql^{-1}). The results are from the same sample (with one offset so they can be seen), the sample locations are aligned to the river sediment results (diamond symbol).

It should also be noted that there was a sample point north of the pipeline that returned tritium activity above the limit of detection, coinciding with the plume expression seen on the beach. Further samples should be collected north of the pipeline to identify if the tritium plume is present in the river to the full extent of the plume seen at the beach.

When compared with the riverbed survey carried out by Whittaker (1977), reproduced in the previous chapter (Section 2.5.2), tritium activity is seen up to over 250m south of the pipeline, compared with only 150m in 1977 (Figure 2-23). This may of course have been the due to limits of the survey grid, in the case of the 2013 sampling round, the riverbank became too steep with rock batters to be able to attain samples any further downstream, or the water became too deep to dig a riverbed sample by hand. Peak tritium activity levels recorded in the riverbed in the late 1970s (Whittaker, 1977;1978) were 133,570 Bql^{-1} compared with 4051.44 Bql^{-1} in 2013 at a similar distance south of the pipeline. The samples taken in 2013 provide evidence that there is still tritium surfacing at the riverbank and in the riverbed sediments. However, without a grid sampling approach drawing conclusions on the controls is difficult. However, given that the tritium results are in a similar distribution to the survey in 1977, we could propose that the groundwater flow surfacing in the riverbed is driven predominantly by the permeability of the riverbed, or could be tidally controlled given the tritium activities are at the tidal limit, possibly not a coincidence? and also be influenced by river bank storage flow processes?

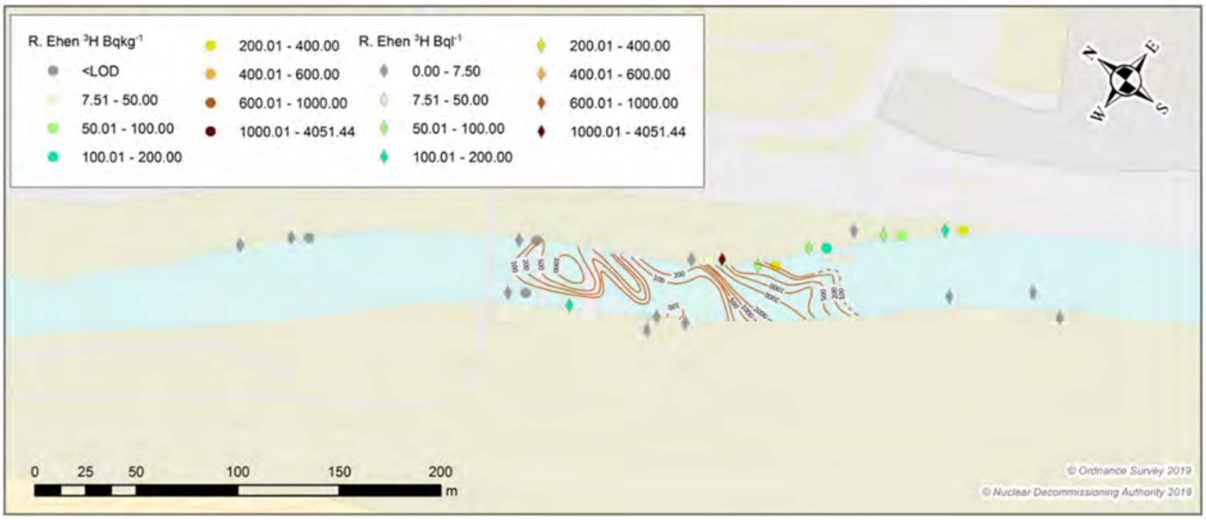


Figure 3-23 Comparison of 1977 river-bed survey contours (Whittaker, 1977) and 2013 river sediment spot samples. The 2013 results for the liquid and sediments are from the same sample (with one offset on the map so they can be seen), the sample locations are aligned to the river sediment results (diamond symbol).

3.3.9 Flow interactions between the groundwater, the beach springs, and the river Ehen.

To explore the relationship between the groundwater travelling towards the coast and the interface with the river Ehen, automatic loggers were placed in groundwater monitoring wells on either side of the river Ehen and results covering the period November 2012 to July 2013 were recorded at 15min intervals (refer to figure 3-3 for locations). The results are presented in Figure 3-24. BH747 and BH6975 from the beach side of the river Ehen are shown. BH747 sits near the river Ehen and within the normal tidal limit and BH6975 is further upstream beyond the pipeline and the normal tidal limit. As previously discussed in the methods section, river levels from a gauging station data 3km upstream were used due to issues with a logger installation closer to the beach groundwater monitoring wells. The results show that the river Ehen water levels generally remain lower than the water table on either side of the river Ehen, but occasionally become higher than the groundwater levels.

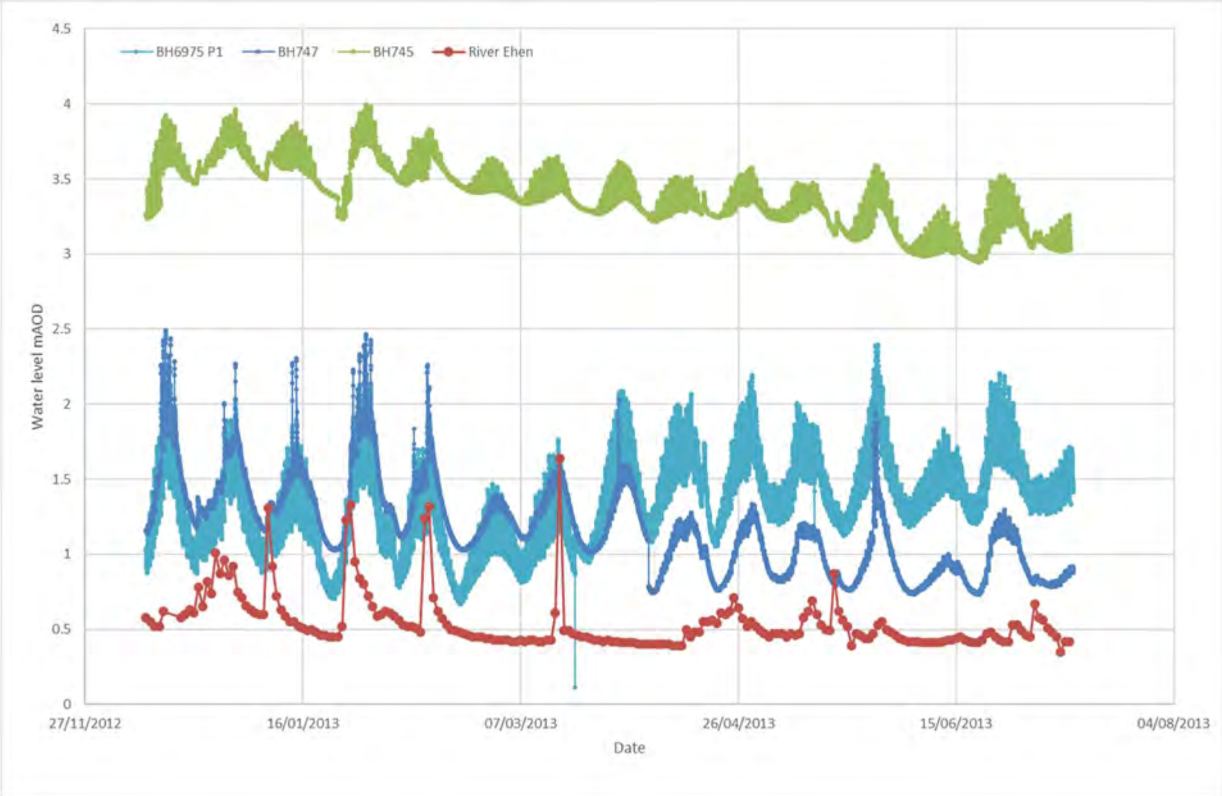


Figure 3-24 Water level fluctuations in groundwater monitoring wells on the west and east bank of the river Ehen, recorded at 15 min intervals and the average river level from the Braystones river gauging station. BH6975P2 is upstream of the normal tidal reach line (NTR) and BH747 is within the tidal reach on the west side of the bank (beach side). BH745 is on the east bank nearest the site perimeter

Figure 3-25 shows a time-period of three months plotted against daily rainfall levels. There appears to be a relationship between periods of higher rainfall and rising groundwater and river levels, though it must be remembered that there will be tidal influences that are not recorded by the upstream river monitoring station.

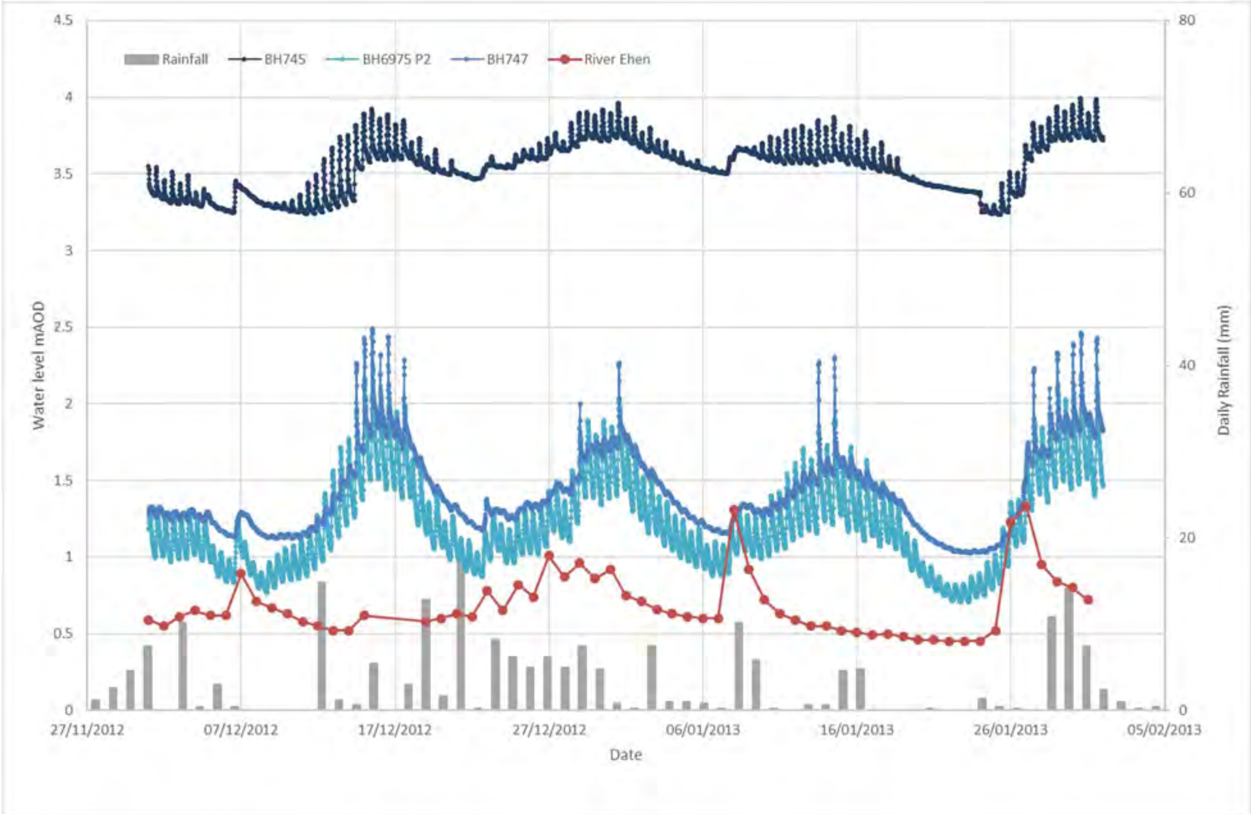


Figure 3-25 Hydrograph of groundwater and river water levels, with daily rainfall levels from the period 1st December 2012 to 31st January 2013.

The change in water levels between the river and groundwater is shown schematically in Figure 3-26. Figure 3-26 (a) shows the relationship of water levels on 7th December 2018, when the river water level was lower than the surrounding groundwater on either side of the river, i.e. the river is gaining during these periods. However, there will be some flow below the river towards the beach. The flows appear to reverse on 8th January (Figure 3-26 b) and are consistent to the observations made in Holmes & Hall (1980) described in the previous chapter. The limitation of these results is that they can only be indicative of general trends as, the tidal impact of river levels is not captured and may influence this flow regime. It is recommended that this experiment is attempted again using automatic water levels loggers in piezometers (surveyed stage and water levels to mAOD properly as one cross section array) to further confirm or disprove the findings of this study.

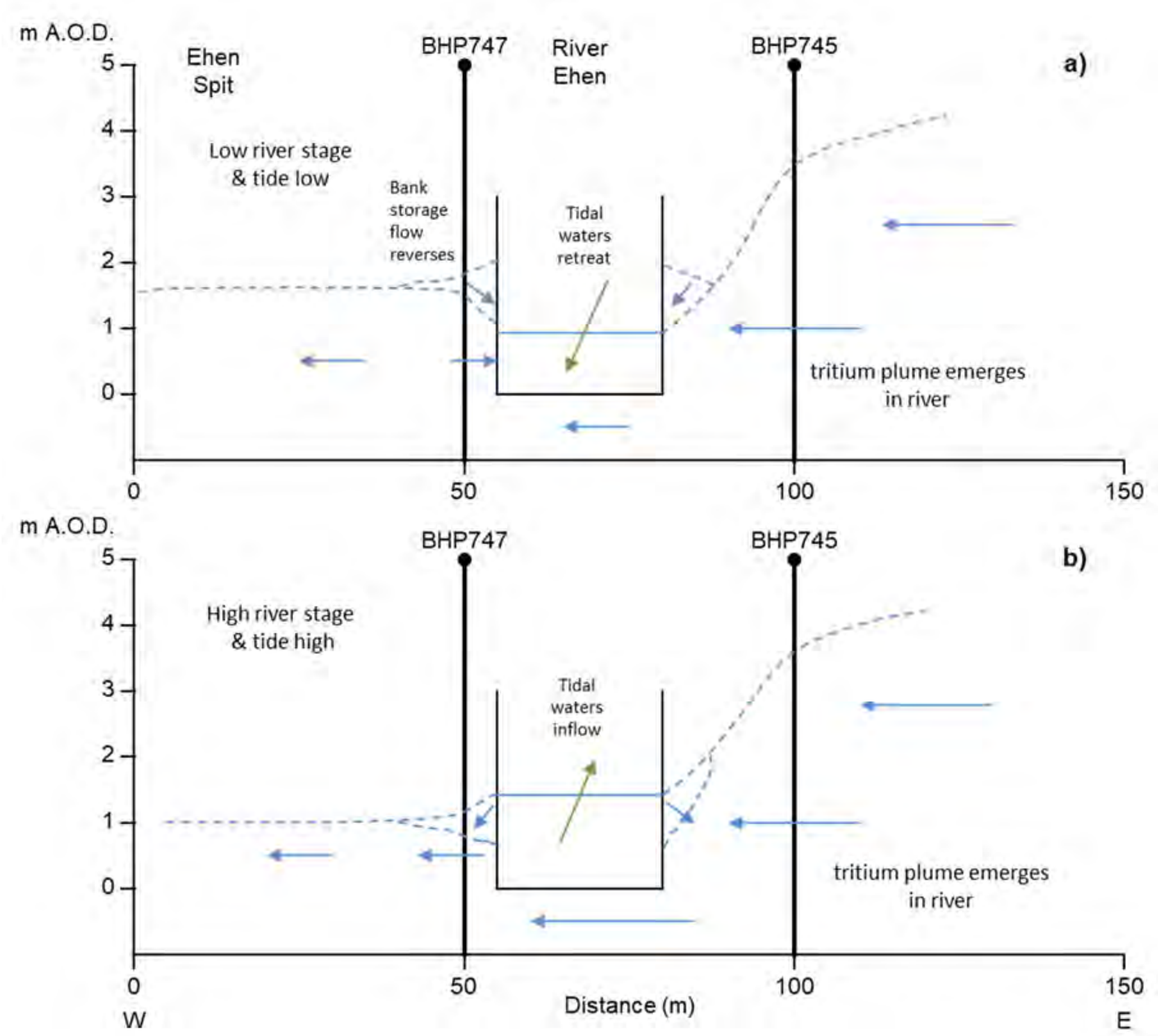


Figure 3-26 Schematic interpretation of the water levels from the hydrographs and tritium sample results a) represents low river stage and tidal waters retreating from the river and b) represents the river stage rising with the tidal water inflowing during high tide.

It is recommended that a further survey of the riverbed is carried out to understand the flows (by measuring changes in head) at this key interface between the riverbed and groundwater. This which would require a small scale drilling rig/mechanical auger and installation of piezometers to avoid the issues of consolidated river bed sediments and the high flow rates seen at times in the river. It could extend a comparable length to the plume expression on the beach. The survey should aim to determine attenuation potential for the radionuclide contaminants (and thereby the natural contribution to risk reduction) as they pass through this hydrological boundary and be captured as part of the Sellafield groundwater conceptual model.

3.4 Conclusions

A comprehensive programme of radionuclide field measurements of beach springs along the intertidal zone near the Sellafield site was carried out from 2009 to 2018. The following conclusions are drawn against the objectives outlines in section 3:1:

1) The results from nearly a decade of beach spring monitoring characterised a distinct laterally stable asymmetric tritium plume discharge footprint of an estimated width of 2600 metres at the beach foreshore.
2) The tritium activity readings in the discharge footprint reduced by approximately half the magnitude over the 10 year interval, consistent with the radioactive half-life for tritium (12.32 years)
3) The lateral extent of the tritium discharge footprint was further north of the Sellafield site than previously predicted by the groundwater monitoring network and the Sellafield conceptual model and may suggest a more complicated groundwater flow regime in the north-westerly reaches of the site.
4) There is one major plume discharge peak at the beach springs, it is quite probable the shoulder and the dispersed tail up the beach are the result of other sources, plumes and pathways.
5) Liquid and sediment samples taken from the river Ehen indicate that the groundwater tritium plume is still surfacing at this boundary in very similar spatial positions to 40 years ago.
6) The long-term temporal profile of tritium activity from 1975 to 2018 shows a steep decline of tritium activity in the first 15 years followed by stabilised low tailing concentrations over four decades. Smaller peaks interpreted as lower magnitude tritium contamination pulses are seen

throughout the time-series results. The temporal record presented is one of the longest globally (over 45 years) of a contaminant discharge profile in groundwater, and possibly the first radionuclide groundwater plume investigated at a beach receptor.

7) Comparisons made with historical investigations summarised in Chapter 2 show good correlation both spatially and temporally with the observed trends from this fieldwork study.
8) This study improves on the earlier investigations (before 2009) with a clear delineation of the spatial plume extent in a very complex double receptor system (river and coastal) and findings should be incorporated into the existing Sellafield site conceptual model.

The key conclusion from this study is that the beach spring monitoring is acting to give an expression of the sources from the site. The beach monitoring also provides an inexpensive far-field monitoring tool at the receptor that should not be overlooked and a significantly lower cost than installation of groundwater monitoring wells. This could improve risk assessment of more attenuated migrating radionuclides.

3.5 Recommendations for Further Work

1) It is recommended that the locations of the beach spring clusters within the footprint are used to investigate groundwater flow pathways by particle back tracking in the Sellafield groundwater flow model to inform on future risks of more attenuated migrating radionuclide plumes.
2) The relationship between tritiated groundwater and the river Ehen hyporheic zone should be investigated further by installation of piezometers and sampling carried out along the riverbed to at least the same distance of the beach spring tritium plume expression and the system understanding updated in the Sellafield conceptual model.

3) A more detailed investigation of the beach spring and tidal interactions could be achieved by installation of shallow beach monitoring well (automatic logging) and geophysics survey and integrated with the riverbed-groundwater investigation recommendation above.
4) Producing an accurate discharge rate is challenging given the low flow beach springs. However, installation of piezometers/seepage metres along a defined traverse would be recommended to produce a more representative discharge rate, with regular water quality samples to analyse for tritium activity.
5) A survey of water levels to understand the flow regime at the groundwater-river-beach interface should be repeated and include river water level data loggers in and beyond the normal tidal limit, with the array of loggers 'surveyed' accurately to provide more precise results.

Supplementary Data Appendices:

A3.1: Sampling procedure & risk assessment for the beach springs monitoring study, and example chain of custody form

A3.2: Sampling protocol/plan for the river-beach groundwater flow interactions study

A3.3: Beach springs field measurement data and tritium analysis results

A3.4: River Ehen sediment sample analysis results

A3.5: Data logger processed data results for beach monitoring wells

CHAPTER 4 TECHNETIUM-99 PLUME BEACH DISCHARGE FOOTPRINT AND IMPLICATIONS FOR PREDICTING FUTURE RADIONUCLIDE ARRIVAL

4.1 Introduction

Technetium exists on the Earth due to nuclear weapon production testing and as a product of the nuclear fuel cycle in reactors and reprocessing (Koide & Goldberg, 1985). There is a very small amount present from the spontaneous fission of naturally occurring uranium (Koide & Goldberg, 1985). Technetium was first obtained from the element molybdenum, but it is more widely known as a nuclear fission product of uranium and plutonium. All isotopes of technetium are radioactive, and the most commonly available forms are technetium-99 (^{99}Tc) and technetium-99m (^{99m}Tc, a nuclear isomer of ^{99}Tc used as a medical diagnostic tool). It is known to be relatively mobile in the marine environment (Leonard et al., 1997) implying a low potential for sediment sorption and has been used successfully as a marine tracer (Aarkrog et al., 1997). Under aerobic conditions, technetium-99 is most likely to be present as pertechnetate (TcO_4^-) (Beasley & Lorz, 1986) which is highly soluble in aqueous solutions. Techentium-99 is found at very low concentrations in the air, seawater, soils, and accumulates in plants and some animals (Beasley & Lorz 1986). The behaviour of technetium-99 in soils can depend on several factors. Redox potential, sorption and interaction with humic matter in soils and sediments plays a role in reducing the mobility of technetium-99 and tends to be retained as relatively insoluble $Tc(IV)O_2$ and therefore the potential for biomagnfication in the food chain is a concern (Icenhower et al., 2010; Leonard et al.,2004; McBeth et al., 2007; Jenkinson et al., 2014). This effect in Brown seaweed (*Fucus vesiculosus*) and certain species of fish, crustaceans and molluscs from the Irish Sea is well reported (Jenkinson et al., 2014; Hunt, 1998, Smith et al., 2001) and highlights the potential risk that radionuclides such as technetium-99 can have in the coastal environment.

Technetium-99 decays with a half life of 211,000 years to stable ruthenium-99 emitting beta particles, but no gamma rays. It is the most significant long-lived fission product of uranium fissions, producing the largest fraction of the total long-lived radiation emissions of nuclear waste. Technetium-99 has a fission yield of approximately 6% for thermal neutron fission of uranium-235. An estimated 160 TBq of technetium-99 was released into the environment between 1945 and 1963 by atmospheric nuclear tests (Beasley & Lorz, 1986). In comparison technetium-99 from nuclear reactors released under regulatory authorisation into the environment up to 1986 is estimated to be on the order of 1000 TBq, primarily released following nuclear fuel reprocessing (such as the La Hague and Sellafield plants), most of this was discharged into the sea (McCubbin et al., 2002). In recent years, reprocessing methods have improved to reduce emissions, but as of 2005 the primary release of technetium-99 in the UK into the environment was by the Sellafield site, which released an estimated 550 TBq from 1995 to 1999 into the Irish Sea (Leonard et al., 1997; Kershaw et al., 1999; Hunt et al., 2013). From 2000 onwards the discharge amount has been limited by regulation to 90 TBq per year, and annual releases from Sellafield have been significantly reduced (<10TBq/year) with actual discharges reported as averaging 1.4 TBq per annum. Annual average technetium-99 activity concentrations are reported as <0.03 Bql^{-1} in seawater samples near the Sellafield site (Sellafield, 2016b). WHO health guideline suggest monitoring limits for technetium-99 in drinking water are set at 10 Bql^{-1} (2011).

Studies of technetium-99 groundwater plumes interacting with the coastal environment at the field scale appear more limited and hence represent a research gap. There are some studies mainly associated with industrial nuclear plants that enrich uranium, or research establishments such as the United States Department of Energy Hanford Site (Beals & Hayes, 1995; Oostrom et al., 2016; LaSage et al., 2008) where technetium-99 groundwater plumes interact with surface rivers/streams. A more prominent area of active research is technologies for bioremediation of groundwater contaminated with technetium-99 (Lian et al., 1996; Lloyd et al., 1998; McBeth et al., 2007; Newsome et al., 2017,2019).

Technetium-99 groundwater plumes are often more harmful to the coastal environment than conservative tritium due to the increased risk of bioaccumulation in the food chain, by direct exposure to contaminated waters or remobilisation of activity stored in sediments. It is therefore key to understand whether this radionuclide, known to be present in the groundwater under the Sellafield site has migrated to the beach area. If this can be achieved, the future risks posed by technetium-99 and indeed other, later arriving groundwater contaminants (e.g., strontium-90) can be assessed. Technetium-99 primary source terms at the Sellafield site are from the leaks at the legacy silos (1970-1982) and the sludge storage tanks (1978-2004) and are not thought to be associated with the Windscale Trenches the primary source for the tritium seen at the beach springs (Garrick, 2010). However, technetium-99 is found at most of the known source plant/leak sites.

Water quality samples that were collected from beach spring discharges between 2009 and 2018 (Chapter 3) were also analysed for technetium-99. The chapter presents spatial and temporal technetium-99 beach discharge data, and a groundwater technetium-99 plume discharge footprint is delineated. The aim of the work described is to demonstrate further the value of analysing beach spring discharge concentrations and to determine whether the results are consistent with the interpretation based on the tritium data (Chapter 3). The footprint of both the tritium and technetium-99, alone and combined will inform forensically on the sources contributing pathways taken to update the conceptual model as necessary. The objectives of the field investigation were: (1) to obtain field data characterising the nature and significance of the technetium-99 groundwater plume discharging on the beach; (2) to compare the observed technetium-99 beach discharge plume with that of the tritium plume presented in Chapter 3 to elucidate further the controls at this interface; (3) to evaluate temporal trends; and (4) consider how the observed footprint informs future risks posed from technetium-99 and more attenuated radionuclides.

4.2 Materials and methods

Technetium-99 is mobile in groundwater when in oxidised environmental conditions, and therefore it can be predicted that the hetergenous drift deposits at the Sellafield site allow the technetium-99 to be mobile, with expected Kd values of <1 it can be assumed that it will travel 90% of groundwater velocity compared to tritium (Icenhower et al., 2010). Sampling of technetium-99 in groundwater at the Sellafield site and occasionally at the beach springs started in 2000, and therefore there is a lack of long temporal records to meaningfully interpret trends as of 2018. However, it was deemed valuable to collect enough water sample volume during the beach spring monitoring study to determine whether technetium-99 was surfacing at the foreshore given similar contributing sources to the tritium. A detailed investigation into the geochemical behaviour of technetium-99 at the beach and the River Ehen was outside the scope of this research study but will provide a baseline dataset to support interpretation for this project and more importantly future research studies. The limited sediment grab samples that were taken in the river Ehen did show darkened sediments indicative of reducing conditions which could warrant further investigation as technetium-99 will sorb to sediments under redox conditions and understanding this potential control on technetium-99 mobility cycling at the river receptor would be important (Masters-Waage et al., 2017).

4.2.1 Beach spring monitoring and sampling framework

A field programme of beach spring monitoring and geochemical water quality sampling was carried out routinely from June 2009 to September 2018. The survey design and event sampling approach are explained in section 3.2.1. A total of 405 samples were collected fortnightly for six months in 2009, and then the sampling frequency reduced to quarterly thereafter. The unfiltered one litre bulk spring water samples were overfilled (to remove air), sealed and transported to the local UKAS accredited laboratory at Westlakes Science Park, Cumbria and radiologically tested for technetium-99.

4.2.2 Technetium- 99 sampling and analysis

The spring water sample is conditioned and submitted to inductively coupled plasma mass spectrometry (ICP-MS) to quantify the technetium-99 activity concentration, using its chemical analogue rhenium, as a tracer (QAAM 674, 2013). Prior to 2013 a gas flow proportional counter was used. Tests were carried out to ensure results were comparable. Technetium-99 has a very long half-life of 210,000 years and therefore any decay correction is negligible. Concentrations are expressed in Bql^{-1} and represent the sample activity of technetium-99 found in the sample at the beach face. Further analysis was carried out to determine the freshwater component of the technetium-99 activity based on percentage of seawater in the sample filtrate, described previously in section 3.2.2 (QAAM 76, 2013). For the 2009 to 2018 samples the minimum detectable activity (MDA) ^{99}Tc was 0.06 Bql^{-1}. The data available from the beach spring peak discharge area from 2001 onwards has been incorporated with the new data collected between 2009 and 2018, giving an indication of the temporal profile over the past 18 years at the beach.

4.3 Results and Discussion

4.3.1 Spatial distribution of technetium-99 in Beach Springs: 2009-2018 observations

4.3.1.1 Location variability of beach spring discharges

A preliminary inspection visit to the beach foreshore in 2009 identified relatively low flow beach springs along the gravel front of the foreshore that were characterised by a lateral area or 'seepage' front that congregated into small pools or channels apparent during times of low tide. 405 water quality 'spot' samples were collected along the Sellafield beach 3 km north of the legacy nuclear site between 2009 and 2018 and analysed for technetium-99 at the accredited Westlakes environmental laboratories. The locations of the water samples analysed for technetium-99 are presented in Figure 4-1, along with the associated technetium-99 activities.

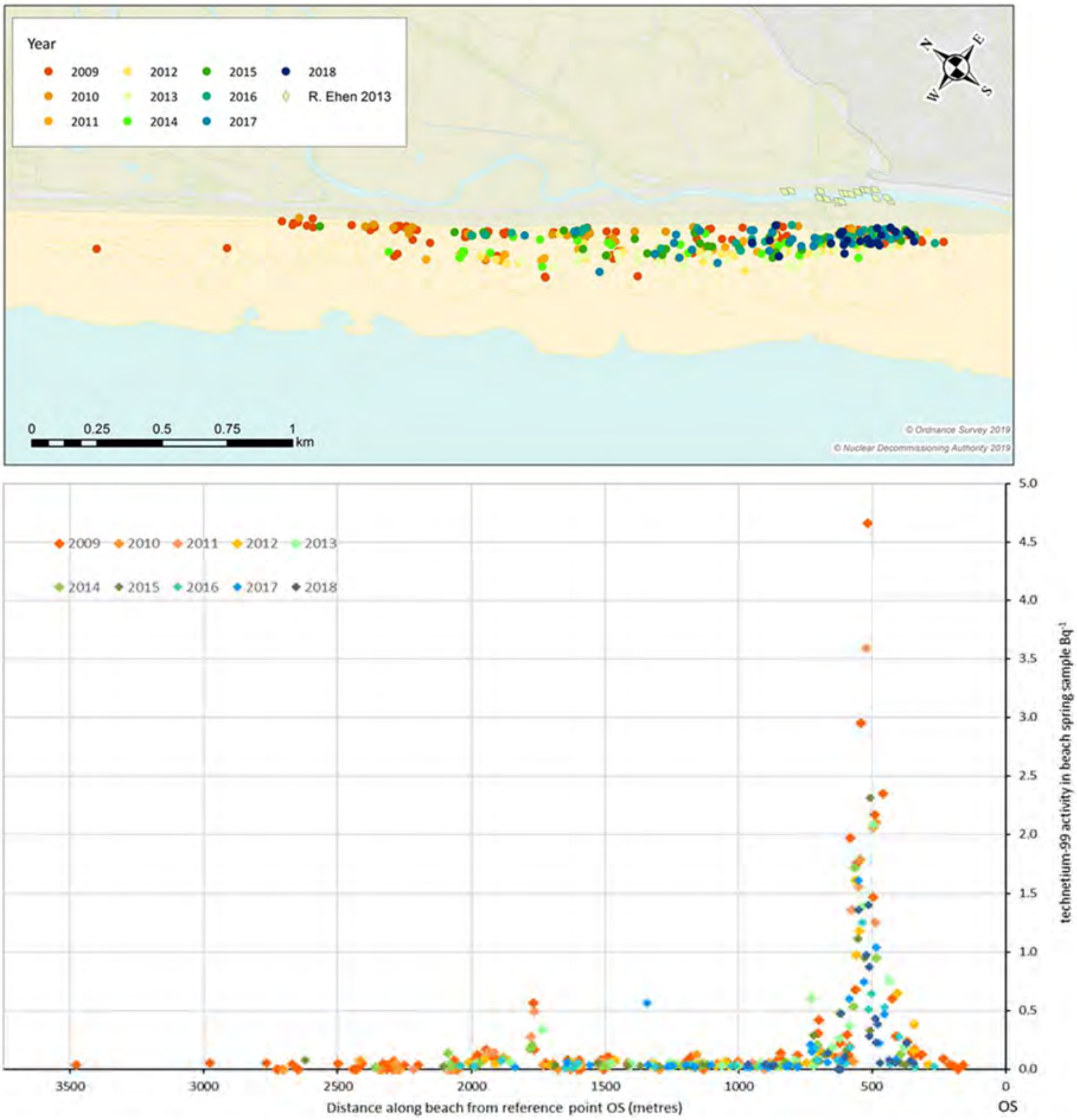


Figure 4-1 Beach spring sample locations recorded GPS locations and technetium-99 activity at each sample point 2009-2018. OS reference point coordinates (zero of x-axis) 30.2267; 50.2835.

Figure 4-2 shows the annual lateral variations and technetium-99 found in spring waters surfacing on the beach for each sample round between 2009 and 2018. The technetium-99 activity appears to remain in the range of 0.1 to 2 Bql^{-1} over the ten years, although two peaks of concentrations over 4 Bql^{-1} are recorded in 2009 and 2011. The peak of the plume discharge footprint aligns to the beach spring peak discharge zone for the tritium footprint (Chapter 3) at around 500 metres along the beach. The data collected in 2018 is not reflective of the full width of the plume footprint, as samples were only taken in the main peak discharge zone area (due to lack of resources) and should therefore not be misinterpreted as sudden reduction in lateral extent.

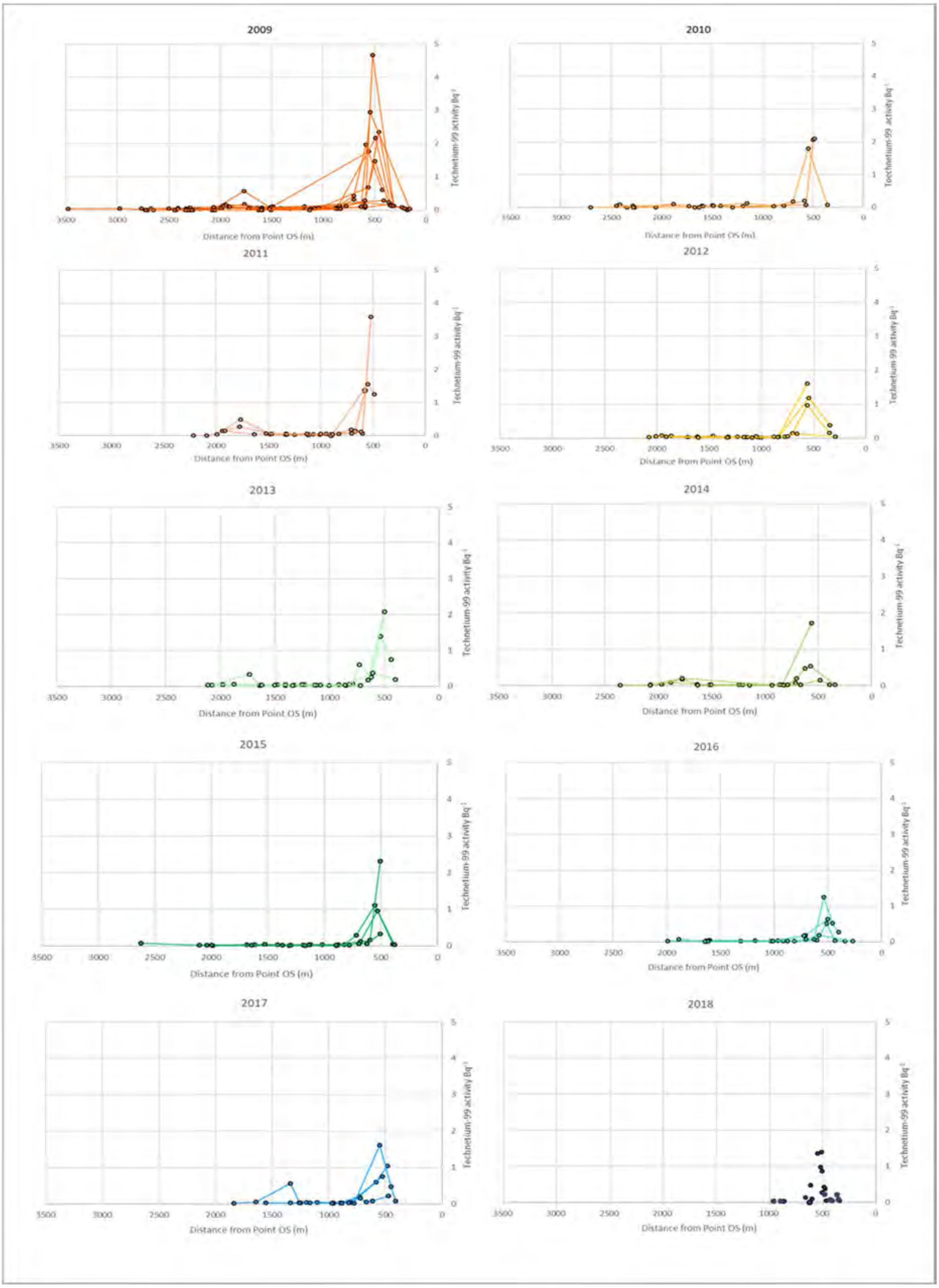


Figure 4-2 Annual lateral variations in beach spring locations and technetium-99 activity (Bql^{-1}) for each sample round collected 2009-2018

4.3.2 Beach foreshore technetium-99 discharge footprint

The technetium-99 activity in the samples collected and analysed delineate a distinct asymmetric groundwater plume discharge footprint stretching to approximately 2600m northwards of the site along the foreshore (Figure 4-3). The beach spring monitoring data have confirmed that the technetium-99 groundwater plume has reached the beach foreshore. The technetium-99 activity levels found at the foreshore are considerably below the WHO drinking water standards of $10Bql^{-1}$ and are of very low risk levels. The background seawater and surface waters (rivers and local tarns) were analysed for technetium-99 and all returned results of below the limit of detection ($<0.06\ Bql^{-1}$).

The plume shape can indicate the type of groundwater flow regime transporting a contaminant to the receptor. The two main source terms of technetium-99 are approximately 1600m and 800m respectively from the coast so if we estimate the aspect ratio (plume length to width) a range of 1.6:1 to 4:1 is calculated, which may indicate a fairly high dispersion of the contaminant to produce the plume characteristics at the beach springs. As previously discussed in Chapter 3 the asymmetric plume shape may be explained by differing source distances, recharge zones, overlapping plumes with different concentrations driven by preferential pathways or by the effects of boundary condition at the river Calder preventing high dispersion of the plume in the southerly direction.

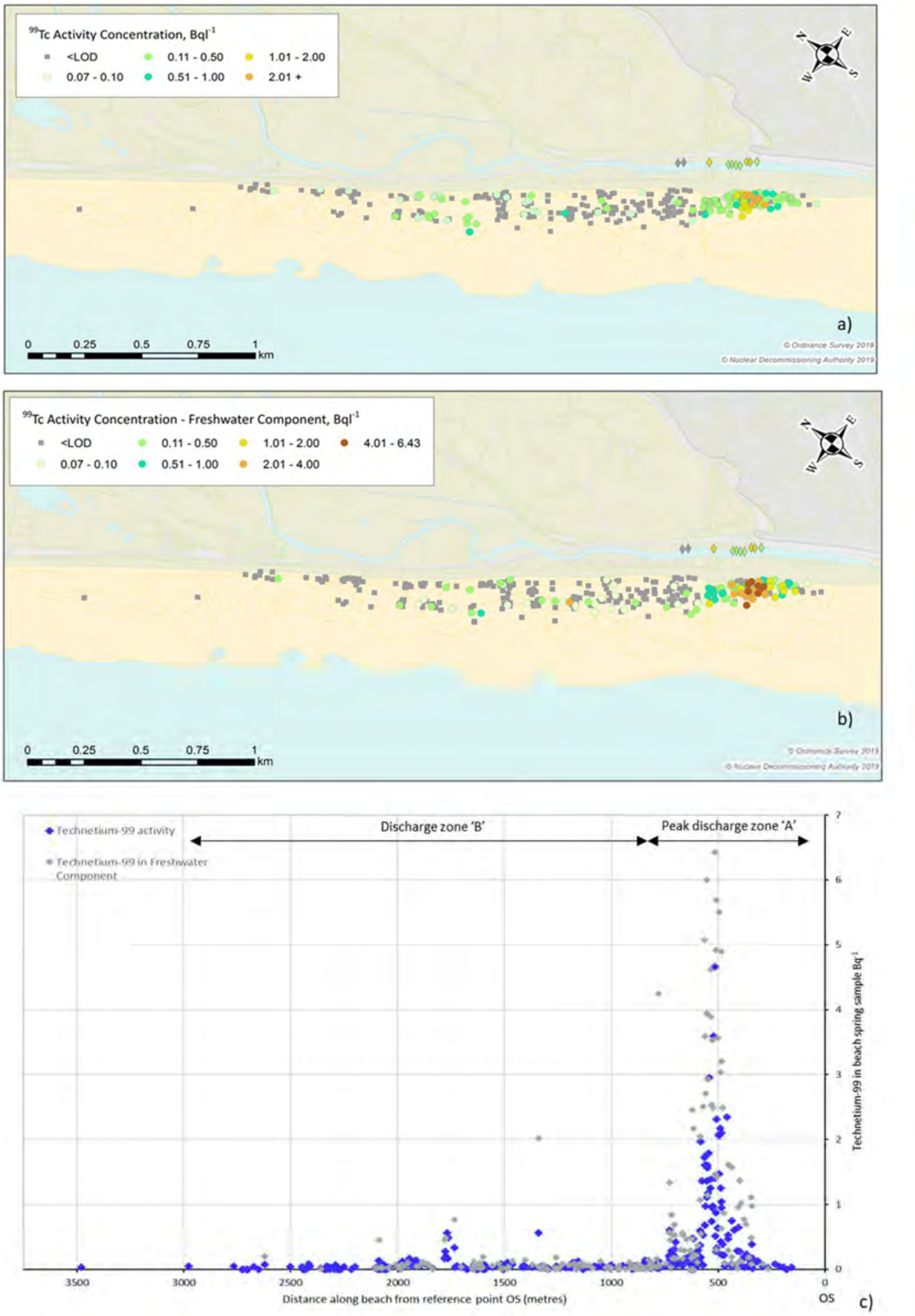


Figure 4-3 Technetium-99 plume discharge footprint 2009-2018. A distinct asymmetric plume of 2600m width, with the spring peak discharge zone area recording the highest ^{99}Tc activity levels, a) ^{99}Tc found in the spring water samples; b) calculated ^{99}Tc in spring water freshwater component; c) ^{99}Tc activity versus distance along foreshore from basepoint 'OS' (30.2267;50.2835).

4.3.2.1 Comparison of tritium and technetium-99 discharge footprints

The discharge footprints for both the tritium and technetium-99 results collected from 2009 to 2018 are presented in Figure 4-4 and show similarities in terms of plume shape and overall width. Both plumes are narrow steep asymmetric peaks and have a lower activity wider shoulder way from the peak northwards.

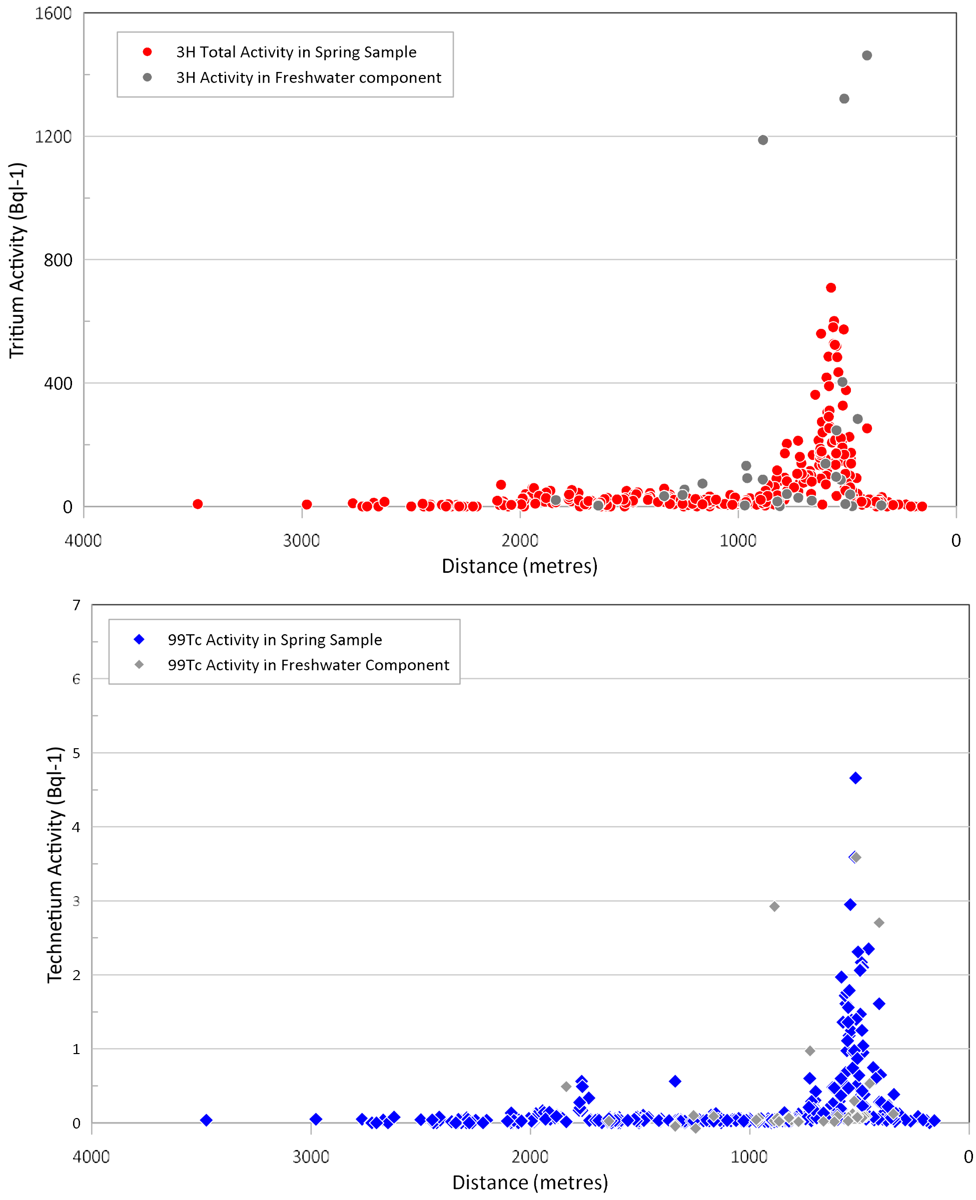


Figure 4-4 Comparison of the (a) sample tritium activity and freshwater component and (b) technetium-99 sample activity and freshwater component against distance (m).

The two groundwater contaminant discharge plumes overlaid on each other appear to coincide spatially, but slightly offset in the peak discharge zone which is interesting as the primary source terms for the tritium and technetium-99 differ on the Sellafield site but are within a few hundred metres of each other. The technetium-99 peak is substantially narrower and more symmetrical than and supports differing source origin but likely on the same pathway trajectory. The consistency in the plume footprints further corroborates the observations made in Chapter 3 around the extent of the groundwater plume to the north-west of the site being further north than predicted from the current Sellafield groundwater monitoring well network and the associated conceptual model. Technetium-99 is predicted to be more retarded (more sorbing) than tritium but appears to have dispersed laterally to the same extent as the tritium plume in the aquifer, so may possibly indicate similar low partition coefficient (Kd) and hence retardation factor values.

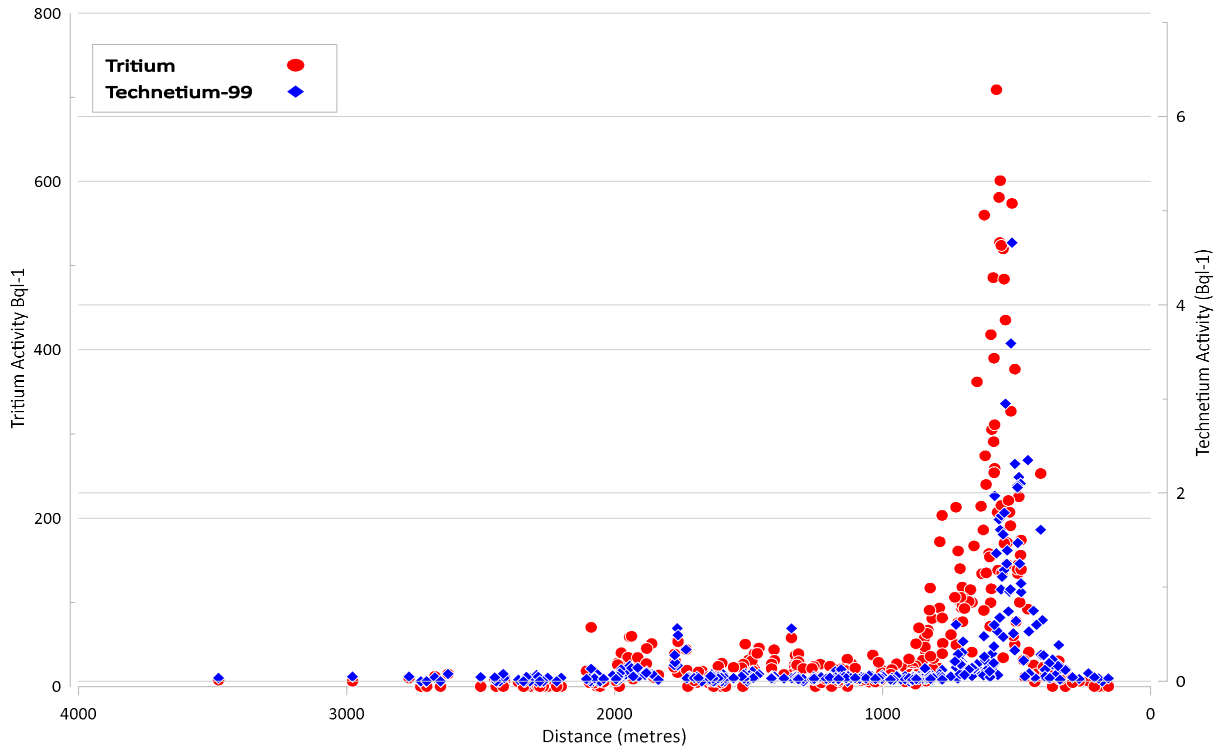


Figure 4-5 Tritium and technetium-99 activity discharge footprint results overlaid versus distance (m)

4.3.2.2 Relationship of tritium to technetium-99 in beach springs

Figure 4-6 plots the relationship between tritium and technetium-99 samples and given the numerous variables in open groundwater systems, there appears two positive correlations possibly indicating two differing sources and mixing of sources between. The results may explain that the highest technetium-99 concentrations are not directly influenced by the peak tritium activity concentrations. This may be attributed to differing source term origins that contribute to the overall contaminant plumes however, spatially they did appear to correlate. An alternative explanation of the low technetium-99 correlation may be related to a pathway with reducing condition that results in the technetium-99 being sorbed. In this case the upper line would represent oxic pathway and the lower line an anoxic pathway.

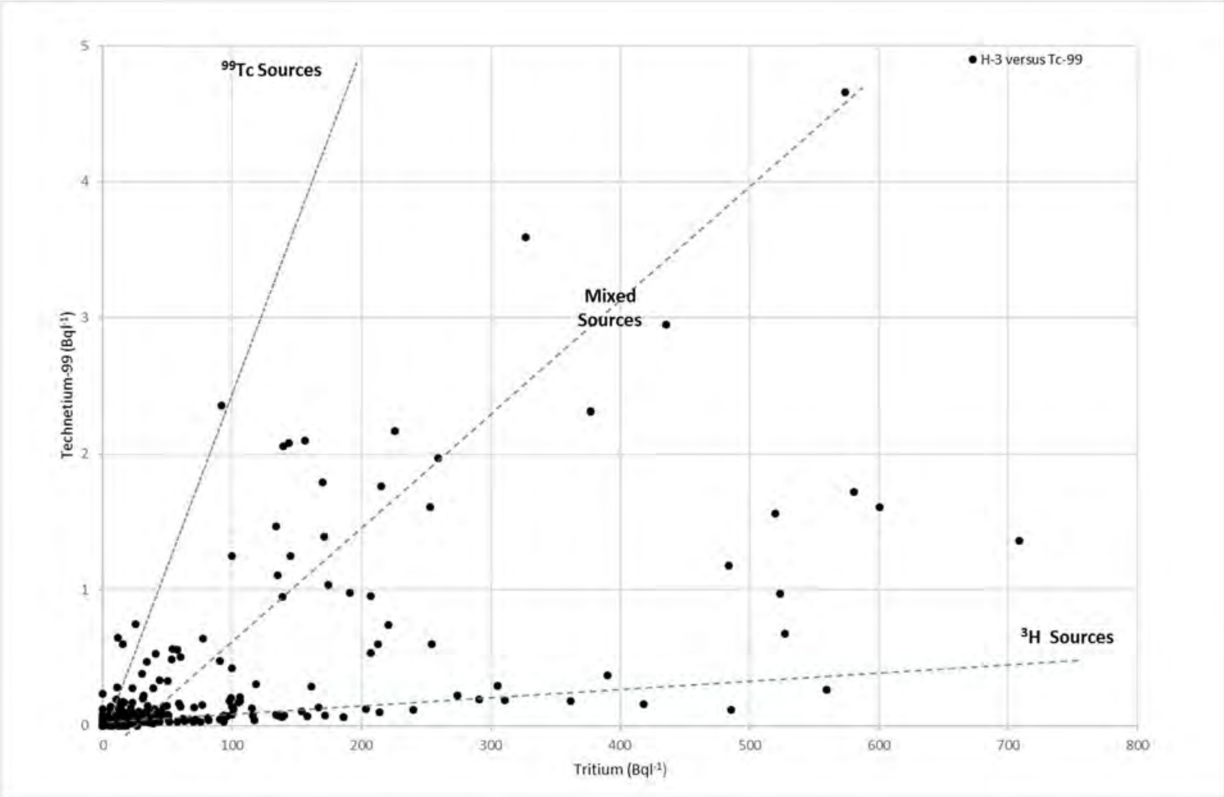


Figure 4-6 Relationship between Technetium-99 and Tritium for samples collected between 2009 and 2018, results may indicate source fingerprints or differing pathway conditions.

The ratio of technetium-99 to tritium is plotted in Figure 4-7 and may provide further insights on this relationship, as the plot shows two groups of distribution across the distance of the beach, the highest ratios tend to be in the peak of the plume, and the lower ratio distribution in the tailing edge of plume. The Windscale trenches (elevated tritium activity) and the sludge storage tanks (elevated technetium-99 activity) are close together (<50m) at the site at a similar distance to the beach of approximately 800m, and therefore the groundwater flow paths maybe similar or merge over a short lateral distances and perhaps indicate a stronger correlation on that basis in the peak of the discharge footprint. Those in the tailing edge may be made up of varying different source terms, with different dilution effects and hence a poorer relationship apart from the merging of contaminants in the groundwater near the beach zone.

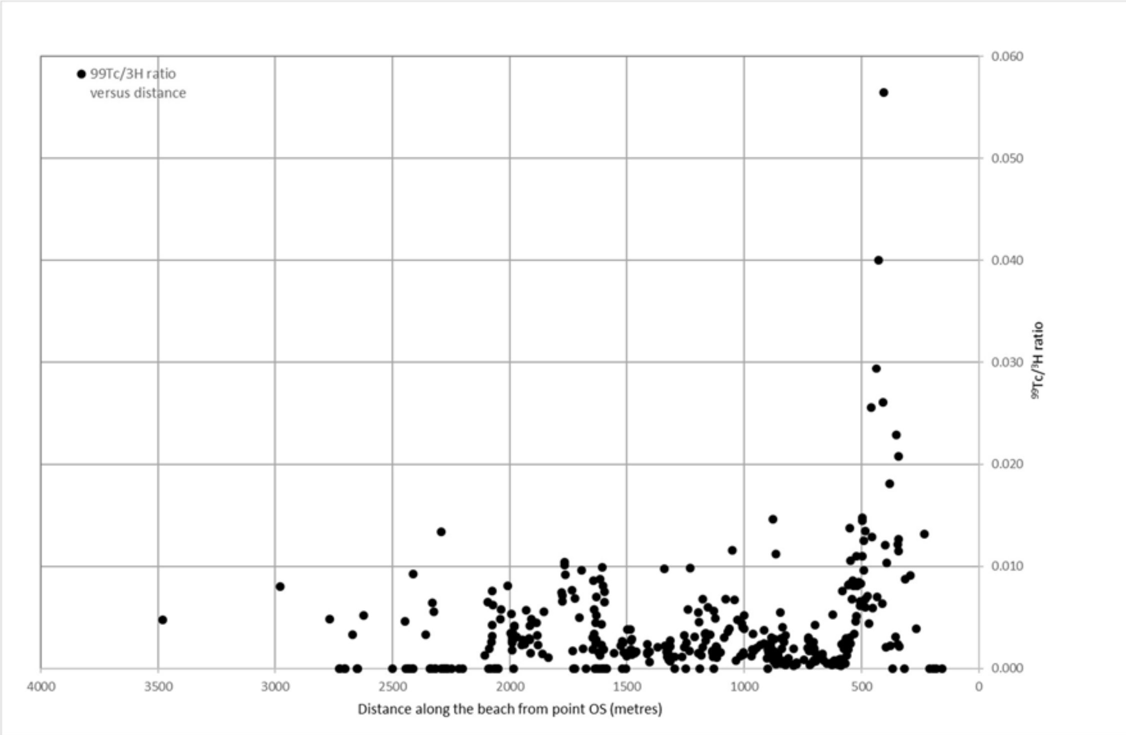


Figure 4-7 Ratio of ^{99}Tc to ^{3}H plotted against distance for samples collected between 2009-2018.

Tritium is present in all the top ten leaks to ground known at the site, and the relationship shown confirms that the tritium has acted as a very good tracer of groundwater transport pathways from the site which appear to merge at the beach springs on the foreshore, all be it a large proportion of contamination will likely be transported in deeper groundwater discharge offshore.

4.3.3 Spatial distribution of technetium-99 in the River Ehen

4.3.3.1 Technetium-99 activity in liquid and sediment samples in River Ehen

Figure 4-8 shows the results of the ‘spot’ samples taken in 2013 along either side of the riverbank. The collected samples were separated for liquid and sediment (dry weight of solids) at the laboratory and analysed for technetium-99. The results should be treated with a little caution as a regular grid sampling was not possible, and therefore the spatial distribution of the samples was determined by the access (depth of water) and collected from areas where the sediments were less consolidated. The maximum recorded technetium-99 activity in the samples was 1.94 Bql^{-1} in the liquid component and 1080 $Bqkg^{-1}$(dry weight) in the sediment component on of a sample on the east bank (Sellafield side). The high points of technetium-99 do not appear to correlate with those of the higher tritium activity in river samples previously presented in section 3.3.8. The considerable amounts of technetium-99 in the sediment samples, may indicate sorption onto the sediments. The total organic carbon fraction recorded for the sediment samples was in the range 0.3-6.6 and the samples with slightly higher levels of the total organic carbon appear to align with the elevated technetium-99 results. It is possible due to the previous large authorised discharges of technetium-99 from the sea pipelines and the sample points being within the normal tidal reach that some technetium-99 may have sorbed from the contaminated seawater flooding the river as the tide rose and circulated and may not all be attributed to contaminated groundwater surfacing in the river water samples, but unlikely given how far offshore the sealines extend (several 100 metres.

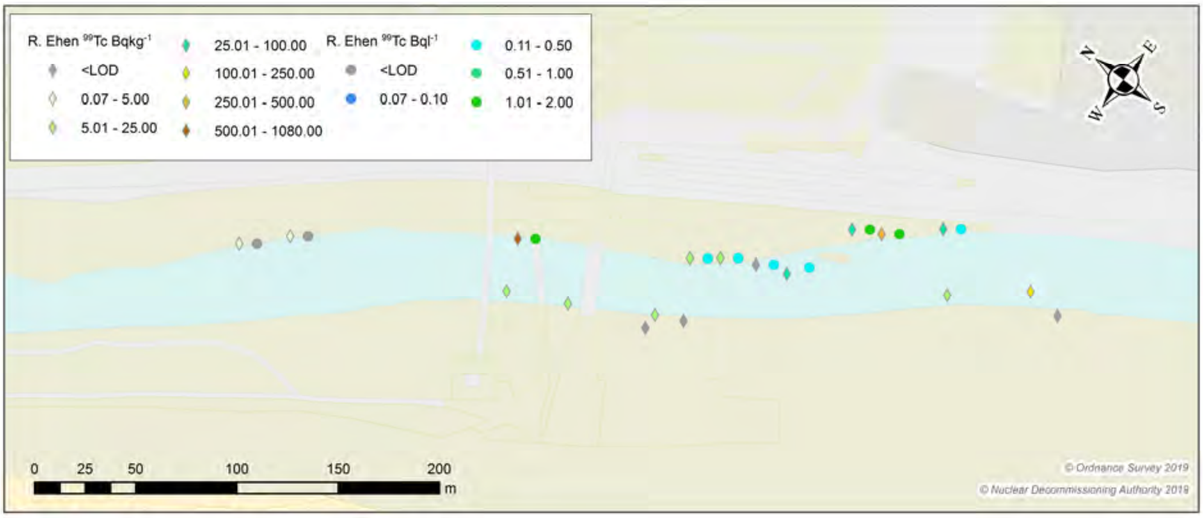


Figure 4-8 Spatial distribution of technetium-99 activity in river sediment ($Bqkg^{-1}$) and liquid samples (Bql^{-1}). The results are from the same sample (with one offset on the map so they can be seen), the sample locations are aligned to the river sediment results (diamond symbol).

There are several sample points north of the pipeline with positive technetium-99 results in sediment samples, coinciding with the plume expression seen on the beach. The samples are beyond the normal tidal limit and therefore would provide confidence that they are related to groundwater surfacing in the riverbed or riverbanks. Further samples should be collected further north of pipeline to understand if technetium-99 is present in the riverbed to the full extent of the discharge footprint seen on the beach. It is recommended that a further survey of the riverbed is carried out to understand this key interface between technetium-99 contaminated groundwater and the riverbed. This will require small scale drilling/mechanical augering and installation of piezometers to cope with the tidal reach and river flow rates present. A grid array is recommended to understand the distribution of technetium-99 particularly within the sediments. A survey of total organic carbon should also be carried out to see if there is a direct relationship between organic content and technetium-99 uptake in the sediments.

4.3.4 Variations in temperature, pH and SPC in relation to technetium-99

Field measurements for temperature, pH, and SPC were recorded for all the sample locations between 2009-2018 and are plotted against technetium-99 in Figure 4-9. Lower SPC readings may correlate with the lower technetium-99 readings in the tailing side of the plume, and the higher technetium-99 results tend to be in the 20-45 $mScm^{-1}$ range. No distinct relationship is seen between pH and temperature against technetium-99 concentrations and are interpreted as seasonal ambient fluctuations similar to that discussed in Chapter 3 for tritium activity results.

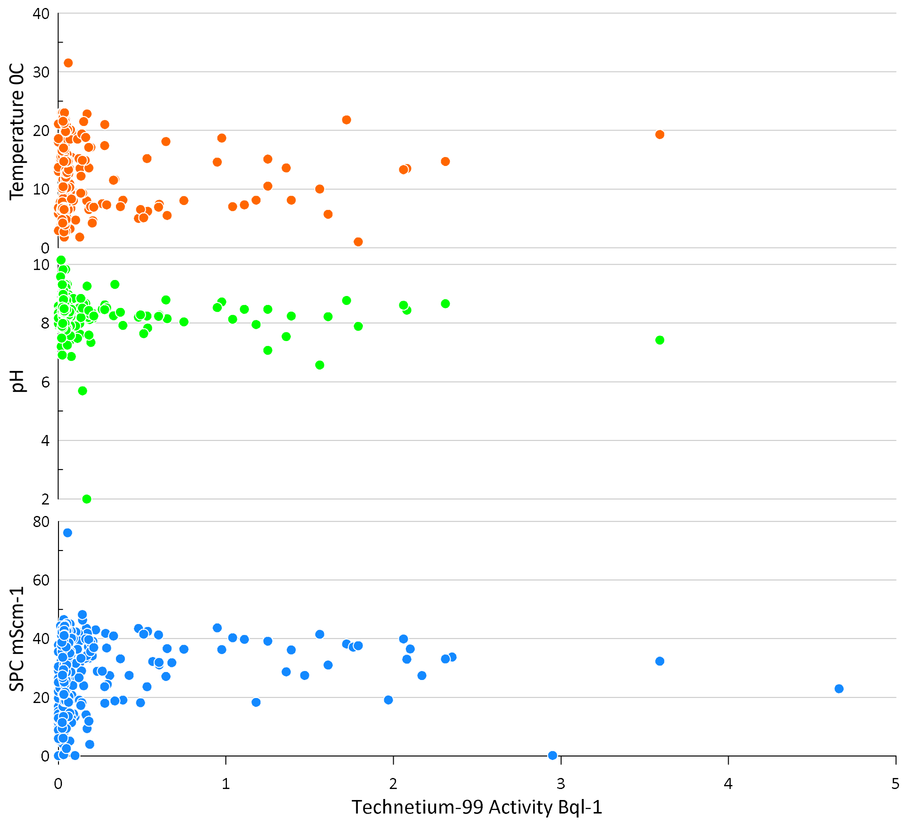


Figure 4-9 Variations of temperature, pH and SPC versus technetium-99 activity 2009-2018.

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4.3.5 Temporal variations of technetium-99 activity in beach springs

4.3.6 Time-series of technetium-99 activity in peak discharge zone 'A' (2009-2018)

Descriptive statistics for the technetium-99 activity samples collected between 2009-2018 are summarised in Table 4-1. Technetium-99 beach spring sample results ranged from non-detect (below the limit of detection of 0.06 Bql^{-1}) to a maximum of 8.75 Bql^{-1} (in freshwater component) across the plume discharge footprint. All the results were below the WHO drinking water standards of 10 Bql^{-1} and considered very low risk to the public.

Table 4-1 Descriptive statistics for beach springs samples collected for the period 2009-2018. Those results that were below the limit of detection (<0.06 Bql^{-1}) are marked '<LOD'.

		Technetium-99 activity (Bql^{-1})				Technetium-99 as freshwater component (Bql^{-1})			
Year	Samples	Max	Min	Mean	St-Dev	Max	Min	Mean	St-Dev
2009	115	4.66	<LOD	0.2266	0.6353	5.43	0.208	2.07	2.91
2010	29	2.1	<LOD	0.2491	0.6032	8.75	0.306	4.425	4.226
2011	36	3.59	<LOD	0.2854	0.6819	2.351	1.644	1.88	0.4039
2012	37	1.61	<LOD	0.1753	0.3533	3.5879	<LOD	0.3748	0.8522
2013	34	2.08	<LOD	0.2112	0.4299	5.5043	<LOD	0.5002	1.1558
2014	30	1.72	<LOD	0.1799	0.3523	5.0739	<LOD	0.6442	1.3412
2015	34	2.310	<LOD	0.1817	0.4479	5.6857	<LOD	0.5041	1.2895
2016	30	1.250	<LOD	0.1502	0.2656	4.6202	<LOD	0.5153	1.0782
2017	31	1.610	<LOD	0.2046	0.3638	4.2476	<LOD	0.5977	1.0775
2018	29	1.400	<LOD	0.3474	0.4324	6.4313	<LOD	1.5135	2.0298

The time-series data for technetium-99 activity collected at the prominent beach spring (peak discharge zone), in the area of peak plume activities are shown in Figure 4-10 for period 2009 to 2018. There is a lack of a clear positive or negative trend in the concentrations over the study period and may indicate quasi steady state plume concentrations or may also depend on how the technetium-99 is being retarded/attenuated along pathways. There are multiple sources on the site and the longer tritium temporal results show the discharges continuing and if we assume comparative mix of sources the technetium-99 is not likely to be declining either. The results are inconclusive in the absence of longer temporal records and needs further long term monitoring, possibly at a higher resolution and frequency to assess the trend further.

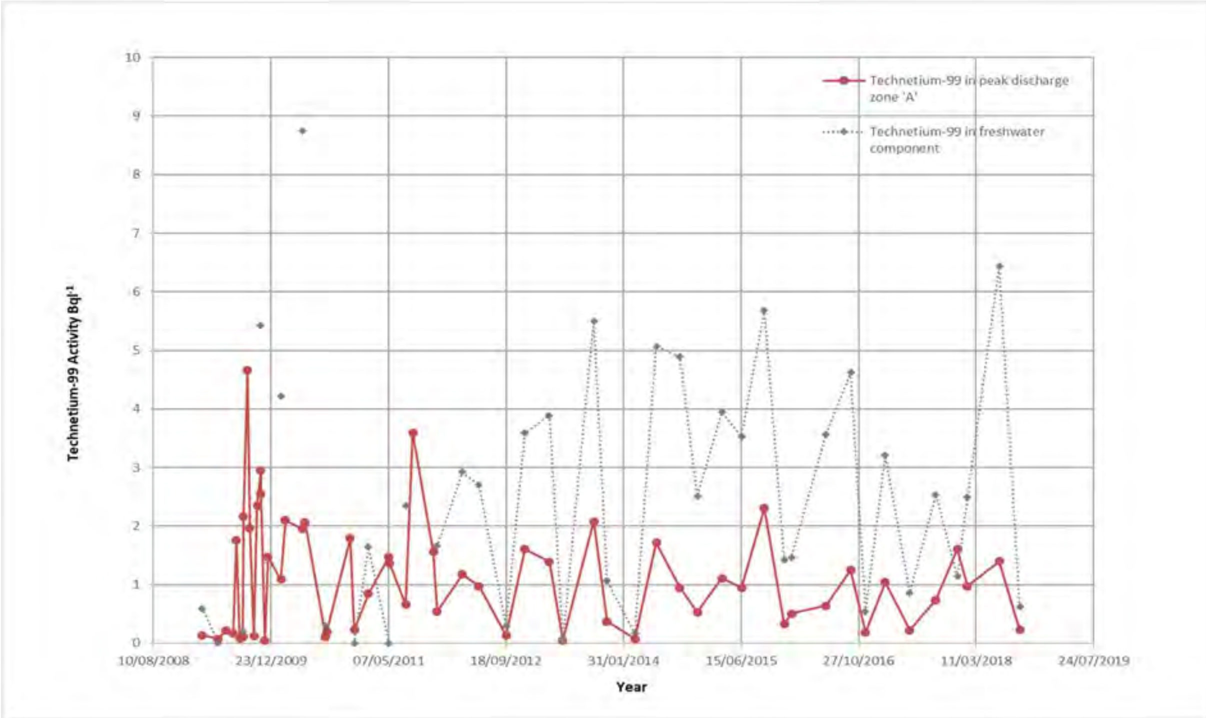

Figure 4-10 Time-series of technetium-99 activity for the peak discharge zone 'A' collected between 2009 and 2018.

4.3.6.1 Temporal profile of technetium-99 in peak discharge zone 'A' (2001-2018)

There are no long-term temporal data sets of technetium-99 at the beach springs that reach back as far the late 70s as is the case for tritium. Some data are available from 2001 in the Sellafield groundwater monitoring database (EAGLE) and are integrated with the new field data in Figure 4-11. There is a data gap between 2004 and 2006 over this longer period of nearly 18 years the trend appears again to be relatively stable. As the technetium-99 is related to similar contributing source terms, and may have similar mobility as the tritium it could be proposed that the peak from those known leaks in the 1970s-80s have already passed through the beach prior to the first samples for technetium-99 at the beach being taken in the early 2000s, if the retardation rates are limited but this is inconclusive from the results. The sludge storage tanks are known to have leaks up to 2004 and likely longer due to residual heels potentially providing an ongoing constant source and therefore the groundwater monitoring wells data for technetium-99 should be reviewed to assess if a peak pulse from that source term is yet to reach the beach, however in the absence of long temporal records the trends would be hard to interpret.

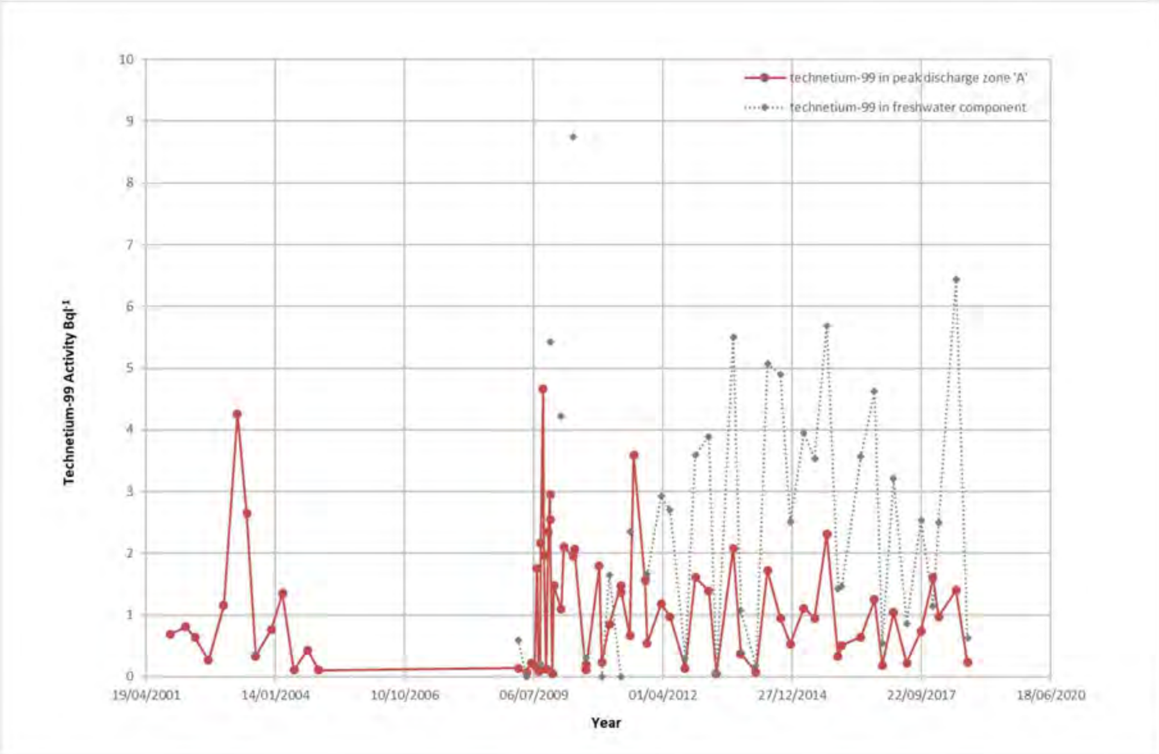

Figure 4-11 Time-series of technetium-99 activity in freshwater component in peak discharge zone 'A' 2001 to 2018.

4.3.6.2 Seasonal fluctuations in technetium-99 activity (2009-2018)

Seasonal fluctuations in Technetium-99 activity for the beach springs has been plotted in Figure 4-12 and for the calculated technetium-99 in freshwater component in Figure 4-13. Spring and autumn are often drier months in this region of the UK and the data collected over the period 2009-2018 show slightly more elevated technetium-99 results in those seasons and suggest that lower technetium-99 activities occur during seasons of higher precipitation (summer and winter) due to greater dilution or dispersion. A similar trend to that seen in the tritium activity data in section 3.3.4. and the same conclusion is drawn that there is not enough resolution of data to provide a convincing seasonal relationship.

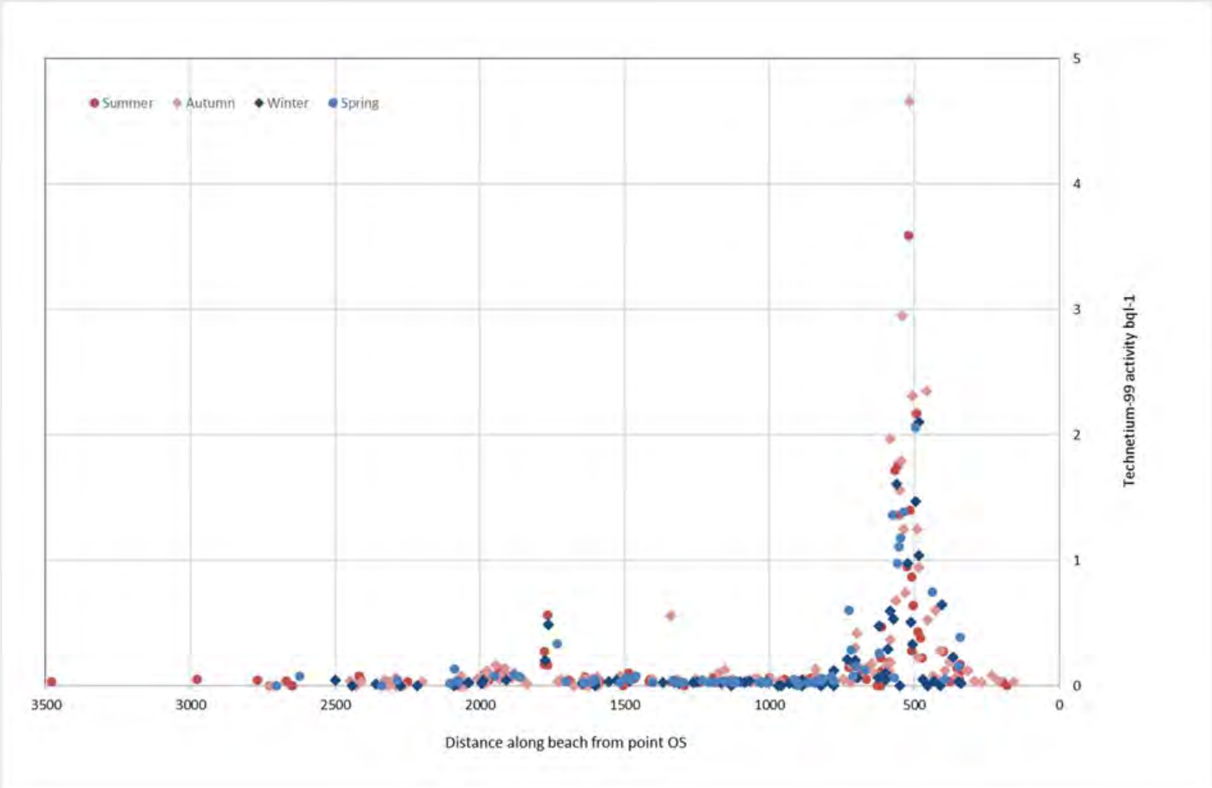

Figure 4-12 Seasonal fluctuations in beach spring technetium-99 activity 2009-2018. Technetium-99 activity is plotted in seasons as Spring (Mar-May); Summer (Jun-Aug); Autumn (Sept-Nov); Winter (Dec-Feb)

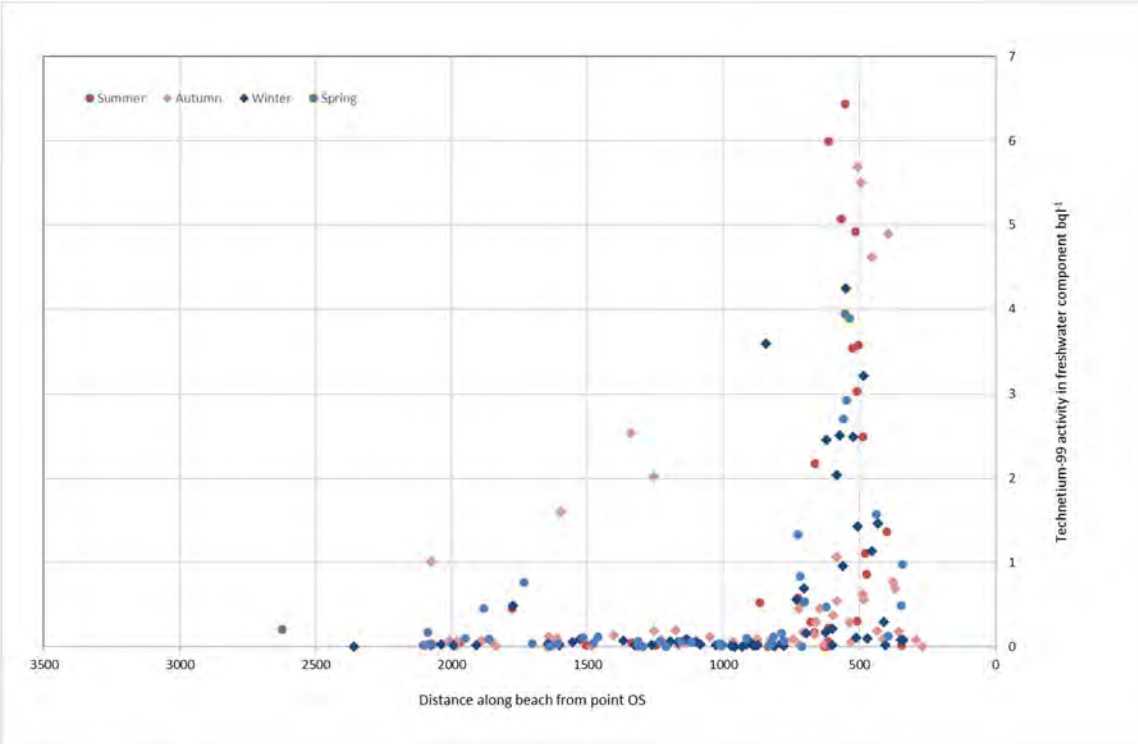

Figure 4-13 Seasonal fluctuations in technetium-99 activity as freshwater component of beach spring water samples 2009-2018. Technetium-99 activity is plotted in the seasons as Spring (Mar-May); Summer (Jun-Aug); Autumn (Sept-Nov); Winter (Dec-Feb).

4.3.6.3 Technetium-99 activity in peak discharge zone 'A' and response to rainfall

Technetium-99 activity and average monthly rainfall are shown in Figure 4-14. The time-series from 2009 to 2016 was analysed so that comparisons could be drawn with the rainfall versus tritium activity profile presented in the previous chapter (section 3.3.4.3). A similar response trend is seen, and technetium-99 concentrations may increase in the spring waters during drier periods, and concentrations reduce during times of higher rainfall with some lag effect evident. The high variability in the most frequently taken data in 2009 points shows the need for higher resolution temporal sampling to explore the relationship further. Taking into consideration other factors such as tides, sampling may need to be a specific event and possibly hourly.

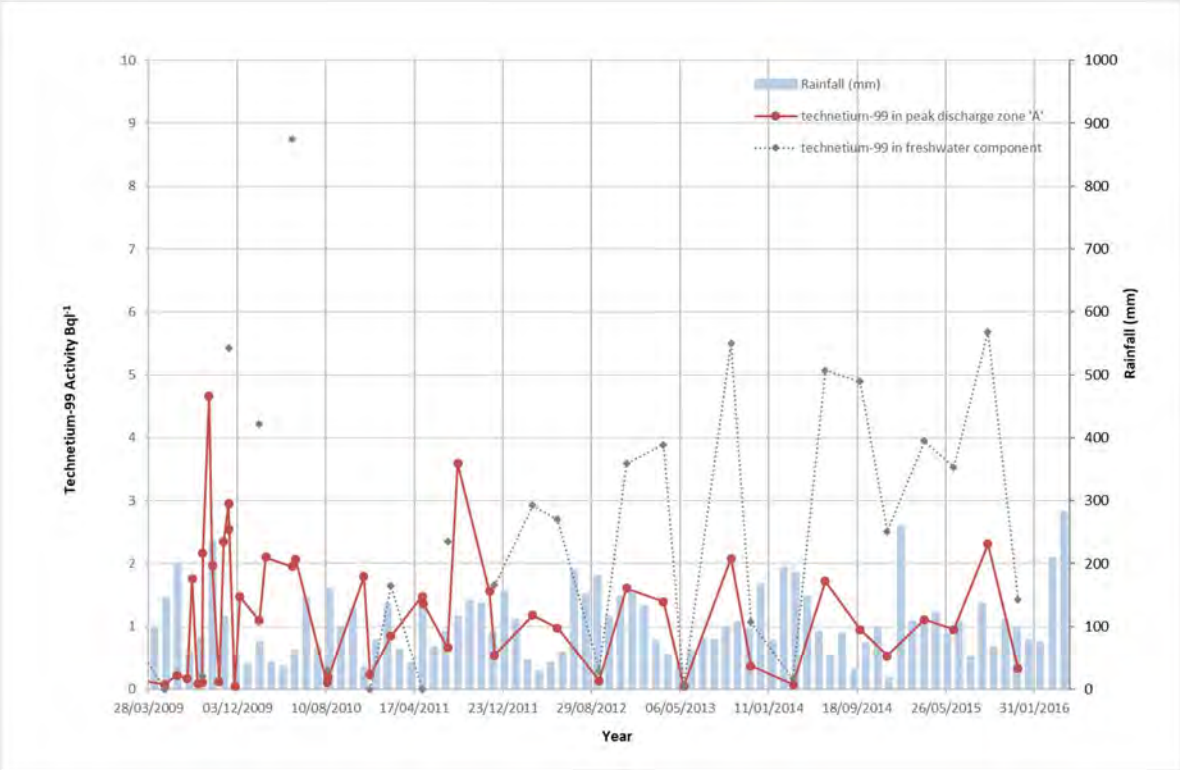

Figure 4-14 Response of technetium-99 to average monthly rainfall fluctuations between 2009 and 2015.

4.3.7 Technetium-99 in beach groundwater monitoring wells.

Long term temporal data for technetium-99 in beach groundwater monitoring wells also does not extend back as far as the 1970s. The more modern 6900 series of monitoring wells do have some records and they are presented in Figure 4-15 for the east and west bank of the river Ehen. Concentrations remain comparable to those find in the peak of the beach spring discharge plume and those on the west side (beach side) could be interpreted as having a rising trend in technetium-99. This is interesting as the tritium levels recorded in the monitoring wells were an order of magnitude higher than the beach springs, yet the technetium-99 levels are comparable. As indicated above there appears to be two sources. The monitoring wells could be showing an increase in plume concentrations at the beach interface, and as such the beach spring discharge concentrations should be monitored regularly to understand if these are also increasing, If this is the case, it might represent a further peak pulse from the sludge storage tanks starting to arrive at the beach. Leakage is known at the sludge storage tanks up until 2004, after this the levels were dropped to heels and given the distance of the tanks from the beach a further peak signature 10-15 years around 2018-2020 would not appear unreasonable. The technetium-99 source from this area is probably ongoing beyond 2004 due to residual contamination in the tanks.

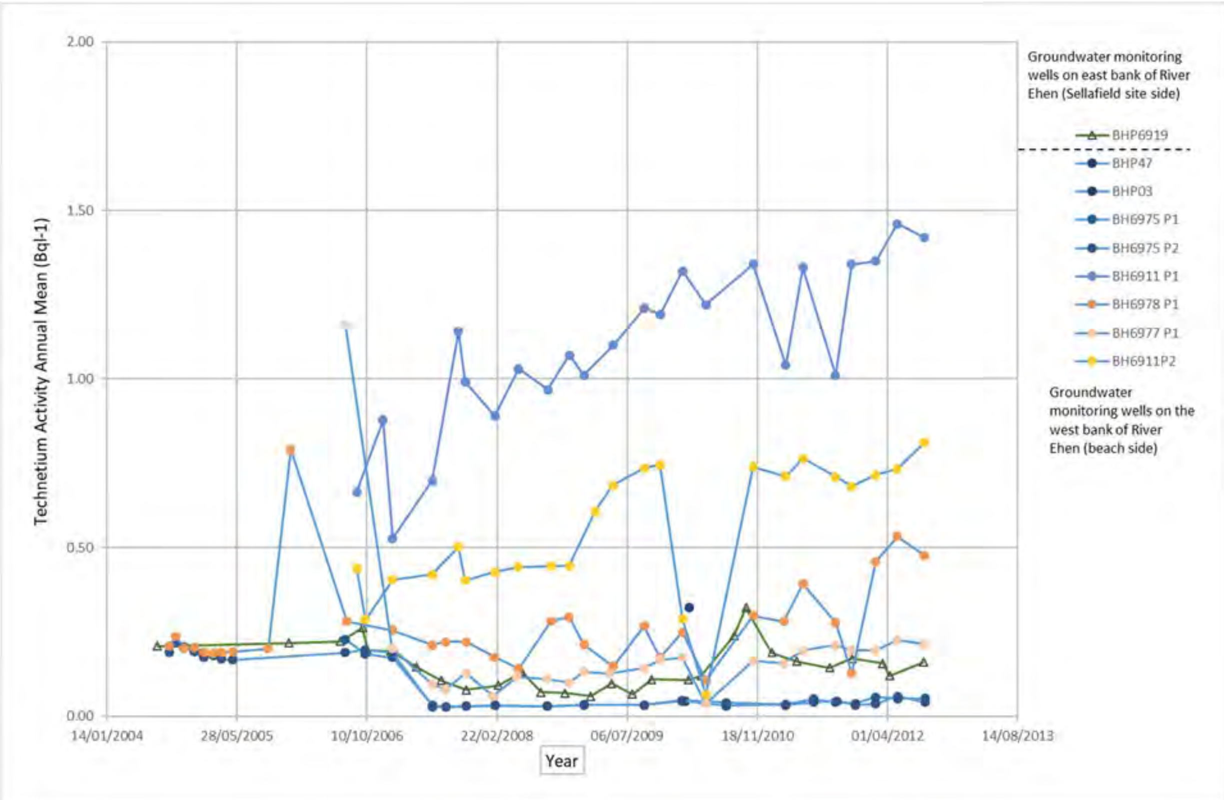

Figure 4-15 Time series profile of technetium-99 activity in coastal groundwater monitoring wells located either side of the river Ehen.

4.3.8 Implications for predicting future radionuclide arrival at the foreshore

The results from the fieldwork presented in Chapters 3 and 4 provide evidence that relatively mobile radionuclide plumes will reach the beach in spring water discharges and thus it can be concluded there is a hydrogeological connection between the source area and foreshore. To date peak discharge zone spring water samples analysed for Strontium-90 have been below the limit of detection (2000-2004), as were the Strontium-90 water quality samples taken from beach groundwater monitoring wells (2004-2012). It should be expected that Strontium-90 will eventually reach the beach. A further field study of the beach springs and the riverbed should be carried out to confirm that Strontium-90 has not reached the beach discharge footprint. To date groundwater risk assessments deem it a low risk to the public and the findings of the fieldwork reported here do not change that conclusion, and the current groundwater plumes appearing to have stabilised at low concentrations, but it should be expected to see occasionally peaks at the discharge from the continuing presence of both tritium and technetium-99 in monitoring wells upstream of the beach. There is a future potential risk to the beach area should an unforeseen or accidental new leak happen at the site as mobile radionuclides will likely migrate along the same pathways as those from the historic leaks. Beach spring monitoring should be continued to monitor trends and the lateral extent of the plume.

4.4 Conclusions

A programme of field measurements of technetium-99 in beach springs along the intertidal zone near the Sellafield site was carried out from 2009 to 2018. The following conclusions are drawn against the objectives outlined in section 4:1:

1) Technetium-99 activity results from nearly 10 years of beach spring monitoring indicate a distinct asymmetric plume discharge footprint at the foreshore of an estimated width of 2600 metres.
2) The lateral extent of the technetium-99 discharge footprint along the beach was further north of the Sellafield site than previously predicted by the groundwater monitoring network and the conceptual model, both of which suggest a more complicated groundwater flow regime in the north-westerly reaches of the site.

3) The discharge footprints for tritium and technetium-99 coincide spatially. Technetium-99 is expected to be more retarded than tritium but appears to have dispersed laterally to the same extent as tritium and it can be concluded that technetium is as mobile as the tritium in this geological setting.
4) Elevated technetium-99 was recorded in both liquid and sediment samples in the River Ehen and may be attributed to contaminated groundwater surfacing in the riverbed and sorbing under redox conditions. Historically technetium-99 was discharged to seawater and may have accumulated from exposure of river sediments to seawater within the tidal reach.
5) Temporal technetium-99 results from the spring peak discharge zone show a lack of distinct positive or negative trend and appear steady state. The historical leaks at the site were mainly in the late 1970s-80s so based on the similar tritium trends seen in Chapter 3 it could be concluded that the peak breakthrough of technetium-99 may have happened prior to the start of technetium-99 sampling in the early 2000's but may be other peaks to follow.
6) The beach monitoring wells technetium-99 show an increasing trend of activity unlike the beach springs and may indicate a further pulse peak of technetium-99 is starting to arrive at the beach.
7) The technetium-99 results from the beach spring monitoring further strengthen the conceptualisation described in Chapter 2 and 3 and the conclusion that the Sellafield site, groundwater, river and the beach springs are dynamically connected. Despite this field study the complexity of this interface warrants further investigation before predictions can be reliably made for the fate of the more attenuated radionuclides.

4.5 Recommendations for Further Work

1) Higher resolution temporal monitoring of technetium-99 is required to understand the short term tidal and seasonal variation of the technetium-99 this will then this will allow the long

term trend to be assessed with more confidence. A longer temporal record is needed than the results of this study both at the beach springs and in the connecting pathways.

2) A survey to monitor Strontium-90 in beach springs should be carried out along the foreshore discharge footprint to definitively conclude that this groundwater plume has not reached the beach to date

3) A riverbed grid survey of technetium-99 activity should be carried out to understand the extent of contamination surfacing within the riverbed and further understand the geochemistry and potential attenuation properties of this interface.

Supplementary Data Appendices:

A4.1: Beach springs field sample data technetium-99 analysis results

A4.2: River Ehen sediment & liquid sample technetium-99 analysis results

CHAPTER 5 CONCEPTUALISATION OF TRITIUM ARRIVAL AT THE BEACH SPRINGS AS AN INDICATOR OF COASTAL RISK

5.1 Introduction

Monitoring of radionuclide contaminant plumes and predicting their future fate are essential for effective management of groundwater contaminants to reduce risk to coastal receptors. The spatial and temporal characteristics of a tritium groundwater plume collated from the archive research study and field observations presented in Chapters 2-4 have provided new insights to the migration of tritium activity to the beach foreshore near the Sellafield site. An interpretation of these investigations is drawn together herein and presented with a further refinement of the preliminary conceptual model proposed in Chapter 2 for the tritium arrival at the beach springs. A long temporal tritium profile at the beach was presented, however is incomplete and reports a partial contaminant breakthrough curve at the beach spring receptor, so characteristics of the plume pathway and travel times to the beach are yet to be fully constrained beyond concluding the primary tritium source was emplaced from 1952 and tritium was found at the beach in December 1975. Due to the conservative nature of tritium it allows the assumption to be made that it 'mimics' the groundwater flow pathways (Plummer et al., 1993; Kendall & Doctor, 2003) and therefore if a theoretical modelled solution can be best matched to the real observed field data at the field scale this understanding will provide some insight into calibrated field parameters and more importantly predict minimum breakthrough times at the beach springs that can then be used as a predictive early indicator of following tritium or other radionuclides plumes that will emerge at the beach receptor.

The aims of this chapter are to (1) assess tritium sources and justify simplifications, (2) interpret the tritium source signature at the beach spring discharge zones, (3) indicate groundwater pathway characteristics using analytical and numerical modelling, (4) collate the technical conclusions from the previous studies (chapters 2-4), and finally (5) summarise the findings in a conceptual model for tritium arrival at the beach receptor near the Sellafield site, and provide recommendations on the priority areas of further research direction.

5.2 Tritium sources at the Sellafield site

The Sellafield site has several sources with the potential to contribute to tritium groundwater contamination within the central area of site which contains many of the legacy operational buildings. All Sellafield reactors have been air or gas cooled and used graphite blocks as moderators. Tritium is produced as a fission product of fuel in reactors with a yield of about 0.01% while some tritium may diffuse through the fuel element cladding and escape into the coolant, most of it will be retained until the fuel is reprocessed and therefore the aerial discharges to the atmosphere were relatively low concentrations during commercial operations with the exception of an accidental aerial emission event caused by the Windscale Pile 1 fire in 1957.

Tritium was also manufactured at Sellafield between 1958 and 1962 (nuclear weapon development) through the irradiation of lithium-magnesium cartridges. During the production period cartridges and associated furnace liners were disposed of in burial trenches or concrete mortuaries located in the north area of the site (operational 1952-1964) and this has been identified in earlier chapters as the primary source of tritium contamination recorded at the beach springs. The primary source term area is located approximately 800m from the beach springs in the coastal zone. The trenches represent potentially the closest source to the coast and one from which significant tritium release was probable and is likely to dominate the discharges found at the beach springs, modelling work herein will therefore focus on this tritium source. The trenches are thought to have been close to the water table at the deepest point of excavation (Figure 5-1). They were firstly open air in the 1950s, covered in a soil mound in the late 1960s, and then capped initially in 1990 to reduce infiltration through the waste, with a further higher specification cap installed in 2014. Tritium will have entered the groundwater from this primary source area as tritiated water (HTO). Some delayed, or phased, release of tritium into the groundwater was probable due to physical containment (wrapping of items in plastic) of wastes with release times being dependent upon rupture induced by overlying waste or ground loading.

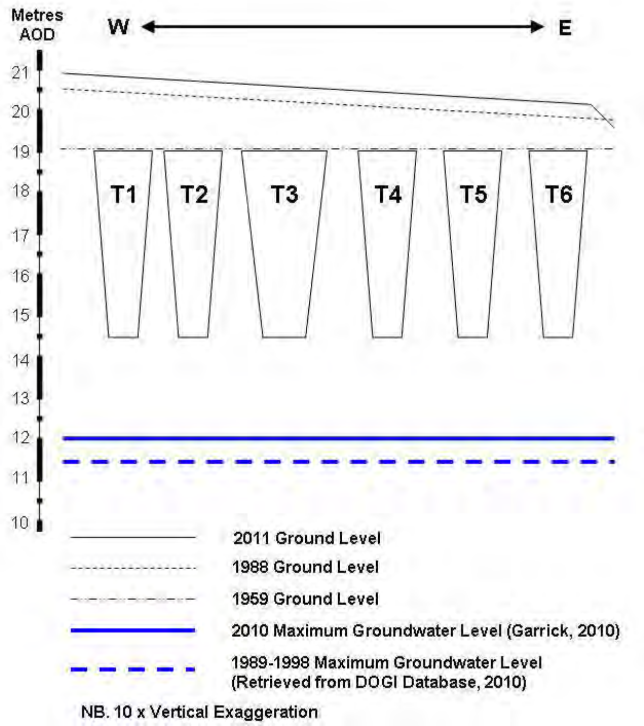

Figure 5-1 Schematic of the primary tritium source (trenches) cross-section and associated capping and groundwater levels.

Other potential sources of tritium of smaller magnitude exist further up hydraulic gradient within the main site area, up to 1700m away from the beach (Serco, 2010). With longer direct travel times to the coast and possibly later release dates, more delayed arrival at the beach receptor is probable and quite likely at a different foreshore location unless happening to lie directly up hydraulic gradient of the trenches source. Less direct groundwater travel pathways to the coast may also further delay and alter the arrival time and location along the beach foreshore.

Most solid waste generated by nuclear fuel reprocessing at the site are stored in various ponds and silos on the site. A small number of leaks from these plants are known and based on groundwater monitoring data in the area are likely to only be small contributors to tritium groundwater contamination and mostly occurred during the early years of site growth, between the 1960s-1980s. Whilst all significant leaks are believed known, there remains the potential for some unknown more minor leaks (e.g. from effluent pipework, or old drains) not easily traced from mass balance or leak-detection approaches.

During fuel reprocessing about 20% of fission product tritium appears in the dissolver off gas and released through the stakes in the gaseous phase. The tritium follows the aqueous phase through effluent treatment facilities to the environment in the low-level waste streams. For many years this liquid waste was handled by a group of ten concrete sludge and settling storage tanks which are located to the south east of the burial trenches and a likely further source of groundwater tritium and technetium-99 contamination from leaks during operations and decommissioning. The final authorised discharge of low-level tritium liquid waste occurs via sea pipelines into the Irish sea.

5.3 Simplification of tritium source characteristics

There is considerable evidence from the studies presented in chapter 2-4 that multiple sources of tritium could possibly be migrating towards the coast, all be it very low concentrations and the tritium activity measured at the beach springs is concluded as very low risk levels to the public. Examining source characteristics at the field scale is complex particularly in the far-field region away from the source origin. The field observations made in Chapter 3 present a unique tritium source signature along the beach discharge zone, and it is therefore deduced that either a wide asymmetric plume expression from one dominant primary tritium source term is arriving at the beach, all be it given the proximity of the source this would require considerable transverse dispersivity over a 800m distance. Alternatively, it is more likely that multiple narrow plumes are merging at the coastal interface, with tritium concentrations controlled by distance from the coast and the highly heterogenous nature of the geology. As a simplification, in the peak discharge zone 'A' it may be controlled by higher concentrations, with sources closer to the beach. The wider shoulder of the plume (discharge zone 'B') is influenced by weaker groundwater concentrations and/or sources further distance away from the beach. Figure 5-2 shows this simplification of the source scenarios and relationship to the tritium beach discharge footprint.

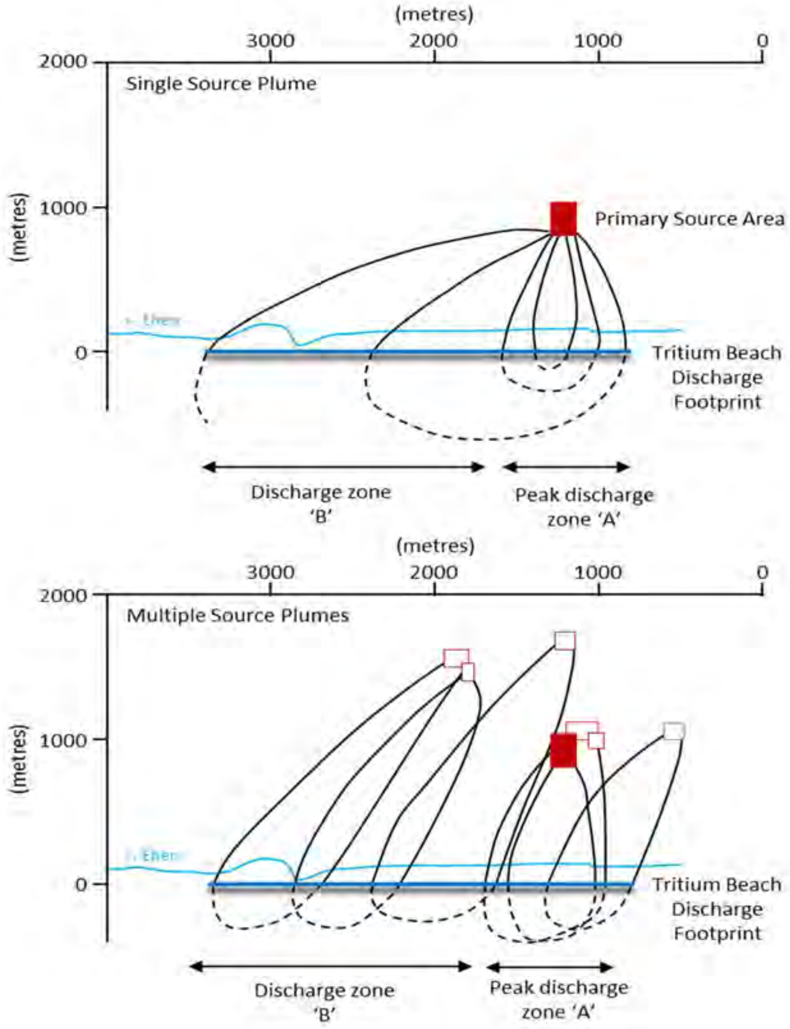

Figure 5-2 Sketch of potential explanations for the high peaks of concentrations seen in discharge zone A, and lower concentrations in discharge zone 'B'. (a) Scenario 1- one primary source creating wide plume (b) Scenario 2- multiple tritium sources creating narrow plumes leading to a merged signature at the beach springs.

The simplified two source scenarios and resultant beach springs discharge plume were further tested using analytical modelling approach described below (Section 5.4). As described previously the trenches (primary tritium source on site) underwent a number of different surface capping regimes, with eventually a engineered cap installed to reduce infiltration in the 1990s, this phasing was explored with analytical and numerical modelling by Rygus (2017) and included the potential for faster preferential pathways in buried channel sediments under the site. The study was unable to achieve a convincing calibration with the field observations, and this was mainly attributed to the high levels of uncertainty around source characteristics and therefore results are not included as part of this interpretation.

5.4 Modelling tritium arrival at the beach springs

Analytical modelling was performed using a one-dimensional transport pathway risk assessment model (Excel spreadsheet) based on the solute transport analytical solutions from Domenico & Schwartz (1997). Analytical modelling is used to make simplifications and assumptions of the flow system to obtain exact mathematical solutions to the solute-transport equation to predict contaminant migration. It is accepted when performing analytical modelling it is expected that some of the complexity of the actual system is lost but allows us to test the conceptualisation principles, to allow prediction of the time window for arrival of tritium at the beach (calibrated with field observations) and importantly identify the key parameters of influence and relative sensitivity. A tiered approach for transport processes was used incorporating dispersion (longitudinal, transverse and vertical), retardation, and degradation. This approach to contaminant transport modelling assumes either a constant source or a finite source during a specified time step and is able too incorporate radioactive decay of tritium.

5.4.1 Model scenarios, assumptions and calibration

The model simulates the release of tritium from the source term area and migration of the contaminant through a homogeneous system at a constant velocity to result in a breakthrough curve of the calculated tritium concentrations at the beach springs. Constant source and finite source models were tested. A constant source was tested to assume that a major leak was constant and ongoing because the beach spring sample results in Chapter 3 showed that when the tritium concentrations were normalised to remove radioactive decay the concentrations appear to decline at first and then remain stable so important to test this scenario. The archive research study however did conclude that the primary tritium source term was caused by solid waste emplaced in trenches between 1952 and 1964 and therefore a finite source contribution is the preferred hypothesis, by assuming that the source is released for a defined time window. It should also be noted that this scoping exercise is approached by inverse thinking, as the field observations at the beach springs receptor gives a footprint of the overall plume(s) signature and therefore part of the solution (dispersion) is given by the dimensions of the plume expressed at the beach. In the absence of long temporal records for the shoulder of the plume (discharge zone 'B') the focus is around the peak discharge area 'A' where records exist for meaningful calibration with field observations. Table 5-1 summarises the four scenarios that were investigated.

Table 5-1 Beach springs contaminant transport scenarios

Scenario	Description
S1: Single tritium plume with constant source	In this scenario it is assumed that there is a constant flux of tritium from a single source (the trenches) to the beach and it is creating the wide asymmetric plume (2600m)
S2: Narrow tritium peak plume with constant primary source	In this scenario is it assumed that there is a constant flux of tritium from the primary tritium source (trenches) and a narrow plume (400m) is created which depicts the peak discharge zone 'A'.
S3: Single tritium plume with finite source	In this scenario it is assumed that there is a finite flux of tritium from a single source (trenches) to the beach and it is creating the wide asymmetric plume (2600m).
S4: Narrow tritium peak plume (primary) with finite source	In this scenario is it assumed that there is a finite flux of tritium (1952-1964) from the primary tritium source (trenches) and a narrow plume (400m) is created which depicts the peak discharge zone 'A'.

The baseline scenario was the starting assumptions for all scenarios as given in Table 5-2, collated from the archive literature review and field observations. Sensitivity analysis was carried out on all the model scenarios as a trial and error method to 'best fit' the computed tritium concentrations to that of the field measurements in the peak discharge zone (A'). The mean annual tritium time-series field data for calibration is shown in Figure 5-3, aligned to the model start time (zero) of 1952, when the trenches were first commissioned and started to be filled.

Table 5-2 Baseline scenario for model runs

Parameter	Value	Origin
Source width	82m	Site archive drawings
Source length	120m	Site archive drawings
Infiltration	0.083 m/d	Gordon (2013)
Hydraulic conductivity	0.4 m/yr	Gordon (2013)
Hydraulic gradient	0.016	Range of 0.012-0.015 in Serco (2010)
Mixing depth	5m	Groundwater data/drift deposits assumption
Porosity	0.175	Serco (2010)
Dispersivity (longitudinal)	800m	Field data at beach springs, distance to source of 800m at the receptor.
Dispersivity (transverse)	2600m	Field data at beach springs, gives plume width at receptor
Dispersivity (vertical)	1m	Assumption based on Domenico & Schwartz (1997)
Radioactive half-live of tritium	4562 days	12.34 years converted to days
Bulk density	1.65 g/cm^3	Assumption based on standards for consolidated sandstone (Serco, 2010)
Kd	0 l/kg	Conservative nature of tritium assumes the same as normal groundwater flow
Distance to source	800m	Nearest main primary source of tritium (trenches)
Finite source time step	4381 days	Assumption based on trenches being open and filled from 1952-1964 c.12 years
Evaluation time	23376 days	To give a temporal profile of 60 years from 1952.

It should be noted that the source term concentration has been omitted in table 5-2 due to commercial restrictions, however it is based on concentrations within the area of the primary source in 1977, increased in magnitude to take account of radioactive decay and then sensitivity tests ran to calibrate to the field data.

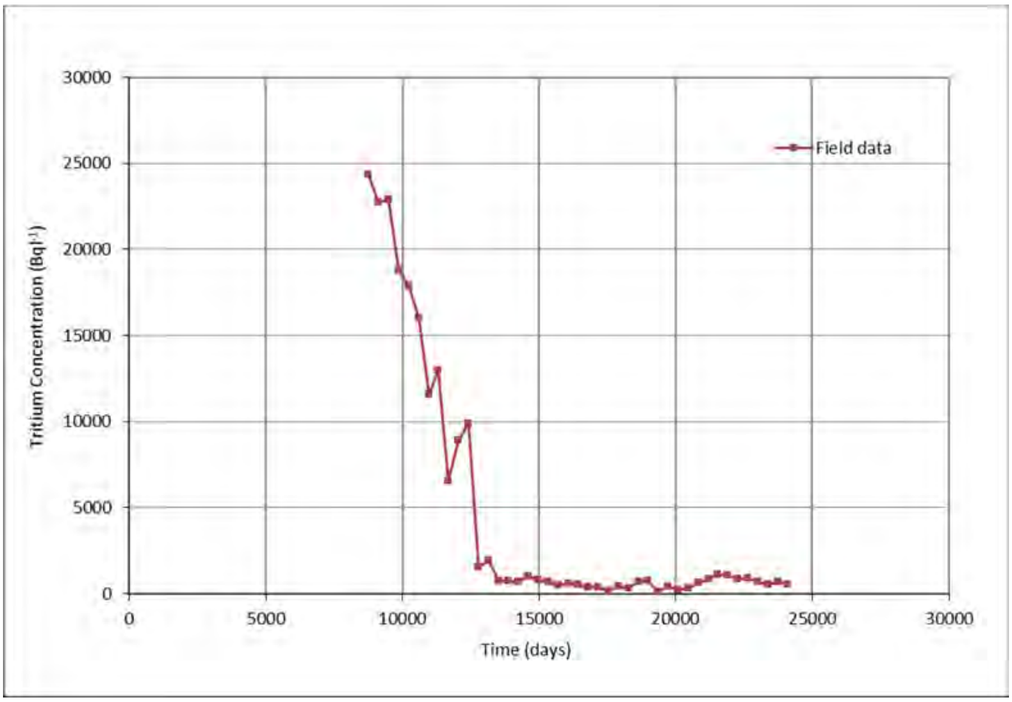

Figure 5-3 Field measurements of tritium concentrations from peak discharge zone ‘A’, aligned to the model start dates of 1952 (when the trenches were commissioned).

5.4.2 Constant source term model results

Scenario 1 and 2 were ran multiple times adjusting the parameters and no good match to the field data was obtained. The results of the two scenarios ran with the baseline parameters is given in Figure 5-4. The source concentration, hydraulic conductivity, hydraulic gradient and the dispersivity were the most sensitive parameters. Scenario 1 and 2 were not taken any further, and it was concluded that a constant source term analytical solution did not provide a convincing calibration to the field observations, and this is possibly explained by the primary source being a ‘finite’ volume of waste placed in the soil rather than an ongoing large plant leak into the ground.

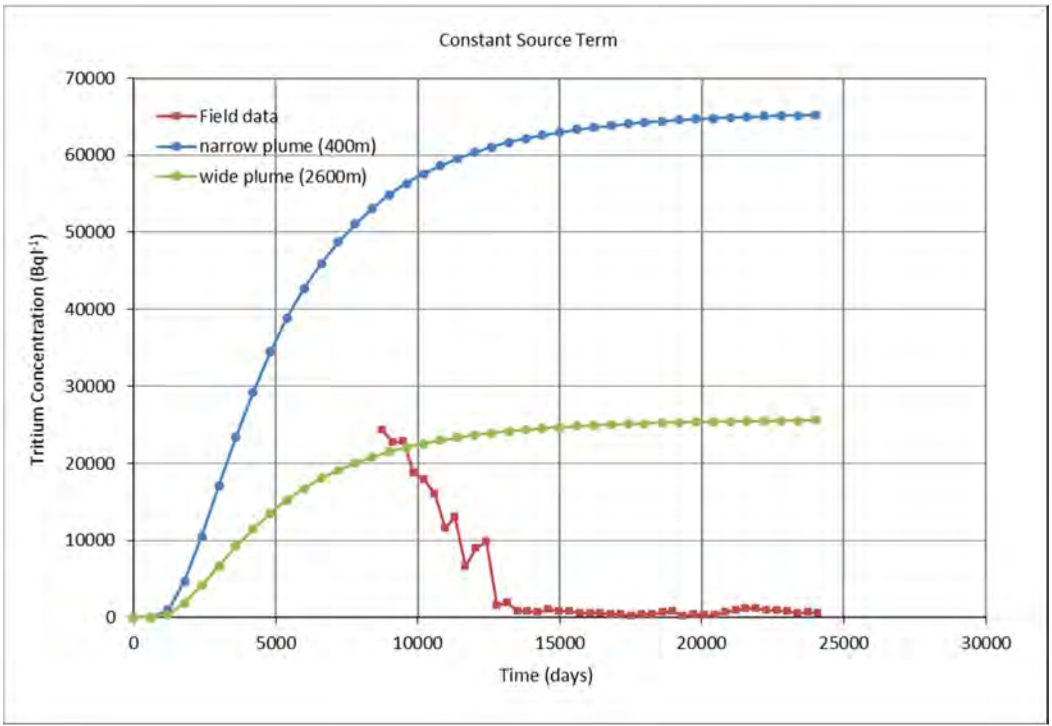

Figure 5-4 Model run results for constant source Scenarios S1 & S2.

5.4.3 Finite source term model results

The results for scenario 3 and 4 model runs are shown in Figure 5-5 and Figure 5-6 respectively, with the associated calibrated parameters. The finite source release of 12 years, and baseline parameters gave the closet result to the observed data. Scenario 4 with a narrow plume hypothesis was the preferred model output as it gave the closest match to the field observations, and an estimated travel time of around 14-16 years to the beach springs. This is lower than that predicted by the Sellafield groundwater model which favours a preferential pathway of around 7-10 years from the trenches to the beach (Serco, 2010). The initial release may have seen the largest flux of tritium enter the groundwater due to flushing when the trenches were open or covered with a light soil cap. The trend seen at the beach looks like a constant source once normalised for radioactive decay (Figure 3.11) but might be explained by some of the tritium waste not have being readily accessible to infiltrating water due to the waste being wrapped or contained in plastics when emplaced in the trenches and later degradation of packaging has then lead to a slow constant release of tritium to the beach springs, which is at stabilised concentrations due to limited infiltration since the engineered capping was installed.

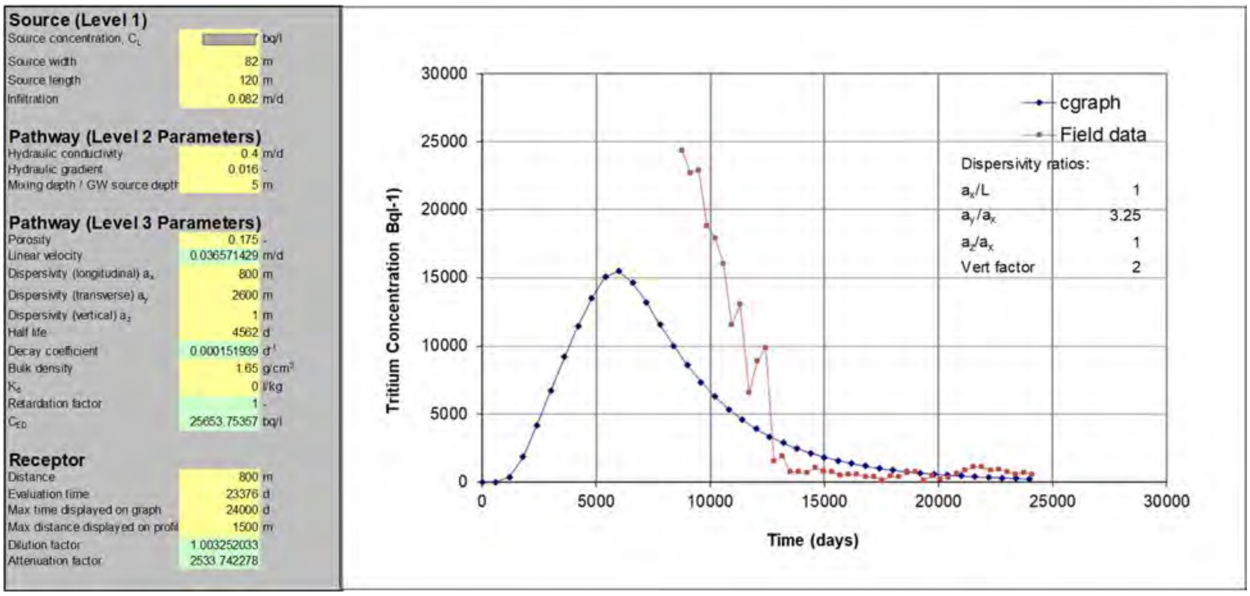

Figure 5-5 Model results for Scenario 3 - single primary tritium source, with wide plume expression at the beach

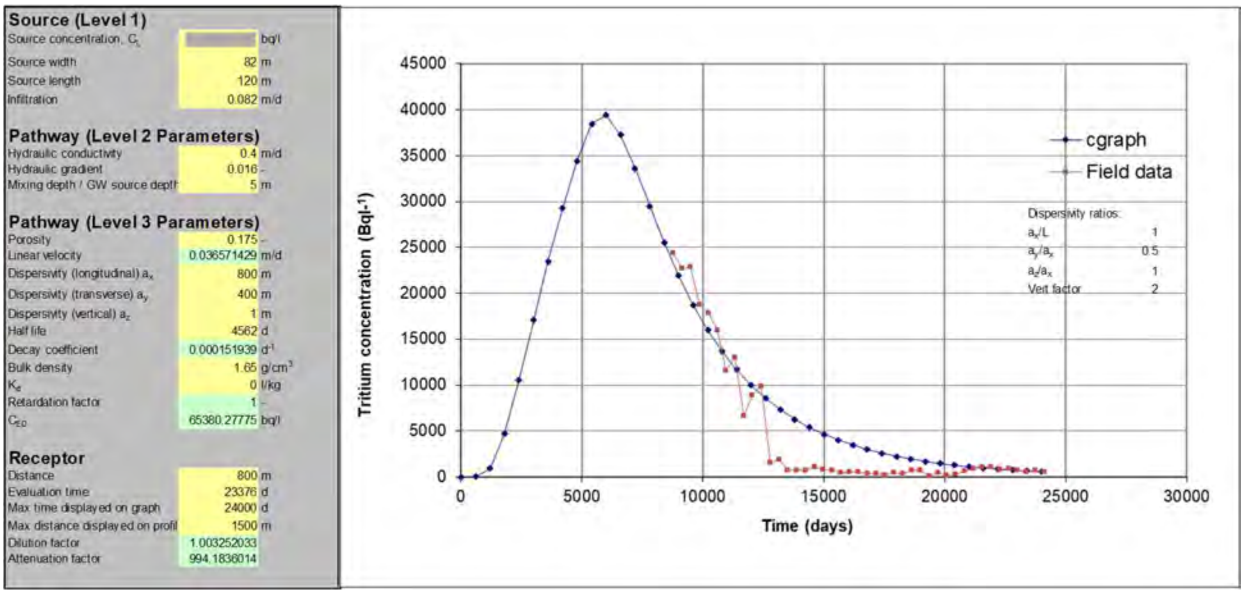

Figure 5-6 Model results for Scenario 4- narrow tritium plume creating peak at the beach spring discharge zone ‘A’.

5.4.4 Sensitive analysis of key parameters

The model results were sensitive to the source term concentrations and was a key parameter when calibrating with field data. The analytical model was calibrated by incrementally altering the following parameters: hydraulic gradient; conductivity; porosity; infiltration rates; source start inventory and mixing depths to match the field observations. The baseline parameters given in table 5-2 and scenario 4 returned the closest match between calculated tritium concentrations and the field observations. Scenario 4 was then taken forwards for further sensitivity analysis. The model runs were particularly sensitive to the dispersivity ratio and the results are shown in Figure 5-7, this was expected as the field observations are taken at the receptor and the width of the plume is known.

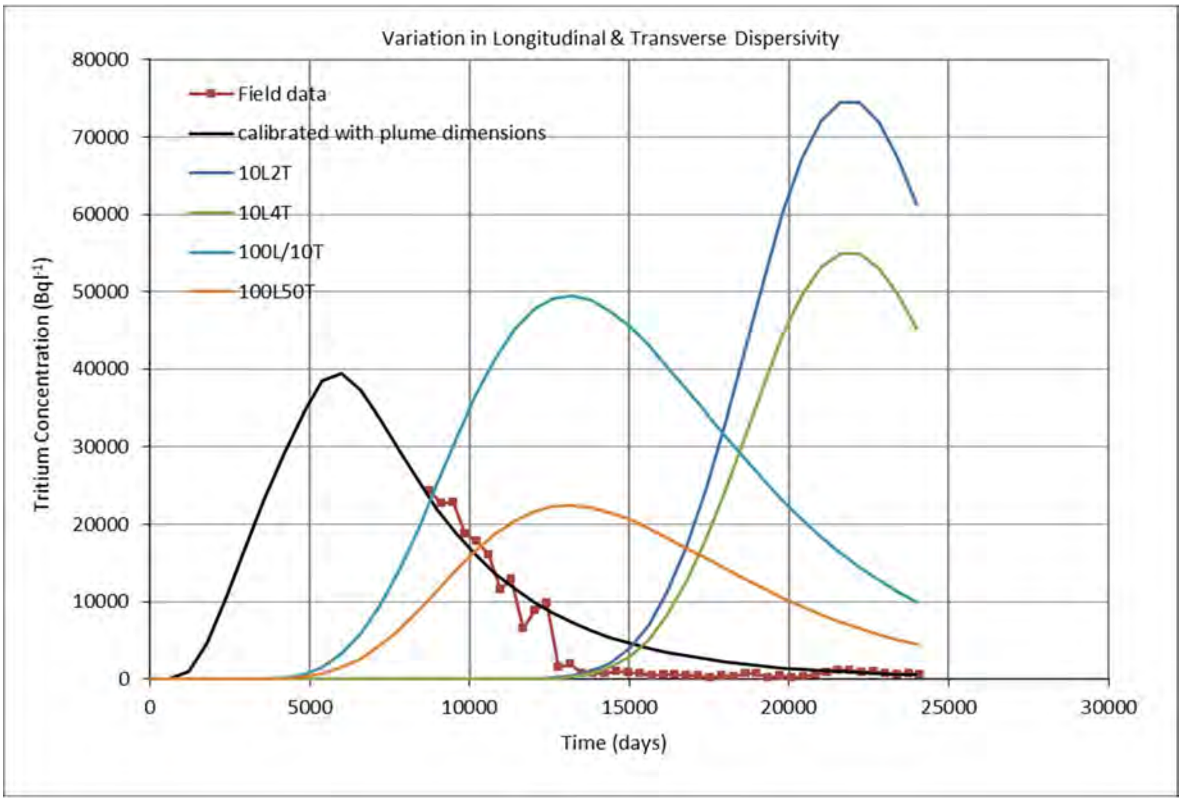

Figure 5-7 Sensitivity analysis model results for longitudinal and transverse dispersivity for Scenario 4.

Figure 5-8 summarises a snapshot of some of the sensitivity analysis runs and the parameters that had most influence on the tritium concentrations. The results were most sensitive to the finite source duration, followed by hydraulic conductivity, hydraulic gradient and porosity. Further refinement of the 'true' field parameters may be required as this model is a simplification of a very complex system. Bulk density and infiltration rates had minimal influence on the results even when large ranges were applied shown in Figure 5-9.

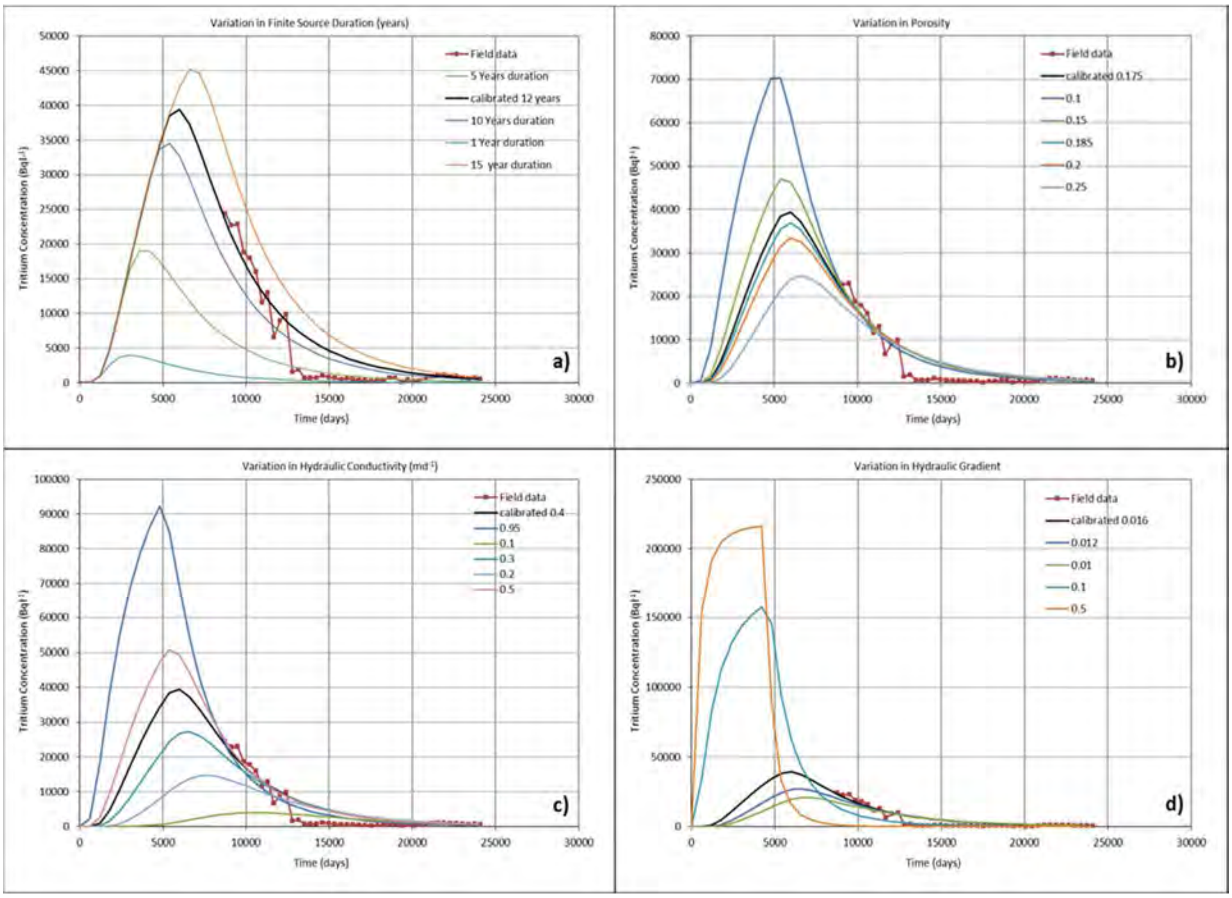

Figure 5-8 Sensitivity analysis for Scenario 4 showing parameters of greatest influence on the model results a) finite source duration, b) porosity, c) hydraulic conductivity and d) hydraulic gradient.

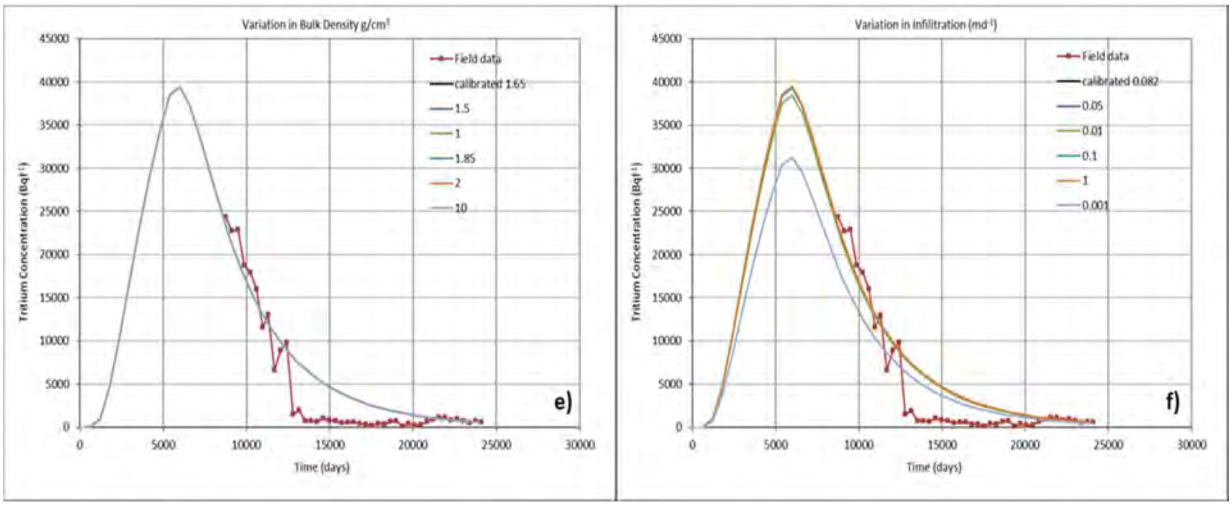

Figure 5-9 Sensitivity analysis for Scenario 4 showing parameters that had minimal influence on the model results e) bulk density and f) infiltration.

5.4.5 Interpretation of the model results

The interpretation of a narrow plumes reaching the beach springs in discharge zone 'A' is further supported by overlaying the multiple source simplification sketch with the tritium discharge footprint in Figure 5-10.

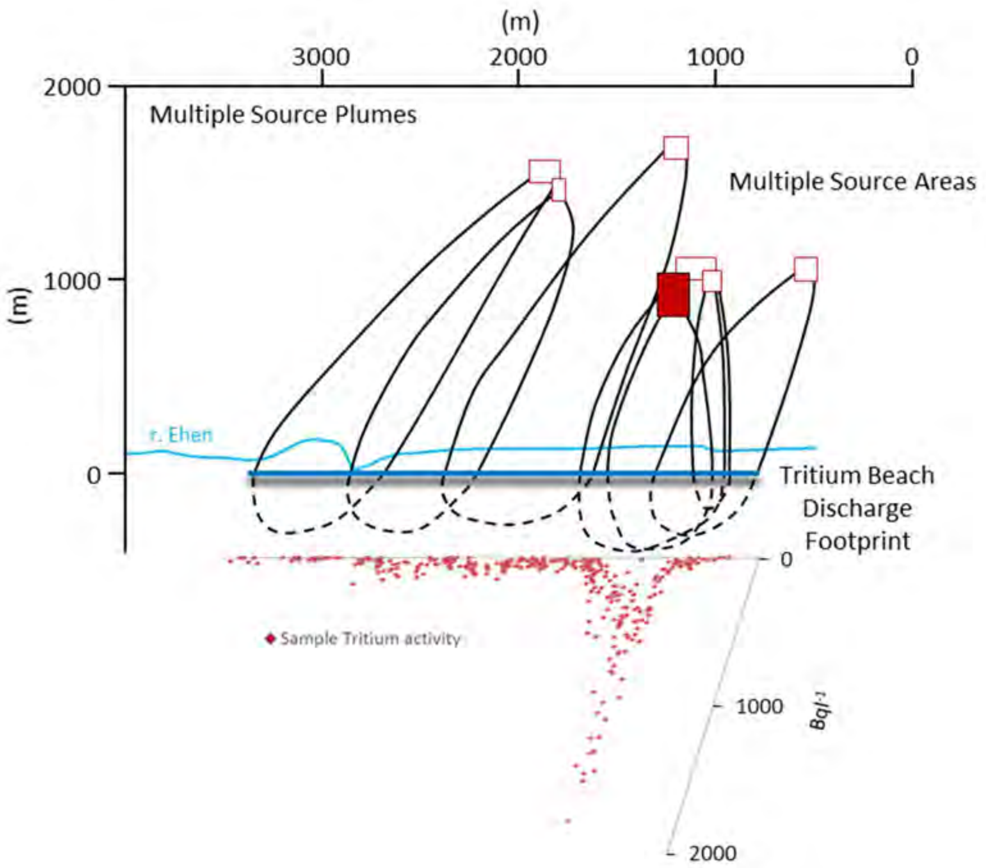

Figure 5-10 Interpretation of multiple tritium source plume expression at the beach spring discharge area.

Technetium-99 is not present in large quantities in the trenches, and interactions would be limited to where upstream groundwater flow pathways intersect the trenches. One of the larger sources of technetium-99 is the sludge storage tanks that are close to the trenches and at a similar distance from the beach springs as the trenches. If we assume that the multiple narrow plume hypothesis is correct and overlay the beach spring technetium-99 results, reasonable agreement is shown in Figure 5-11, and strengthens the conclusion. In the absence of a good temporal profile of technetium-99 to calibrate model results the analysis for technetium-99 was not carried any further in this study.

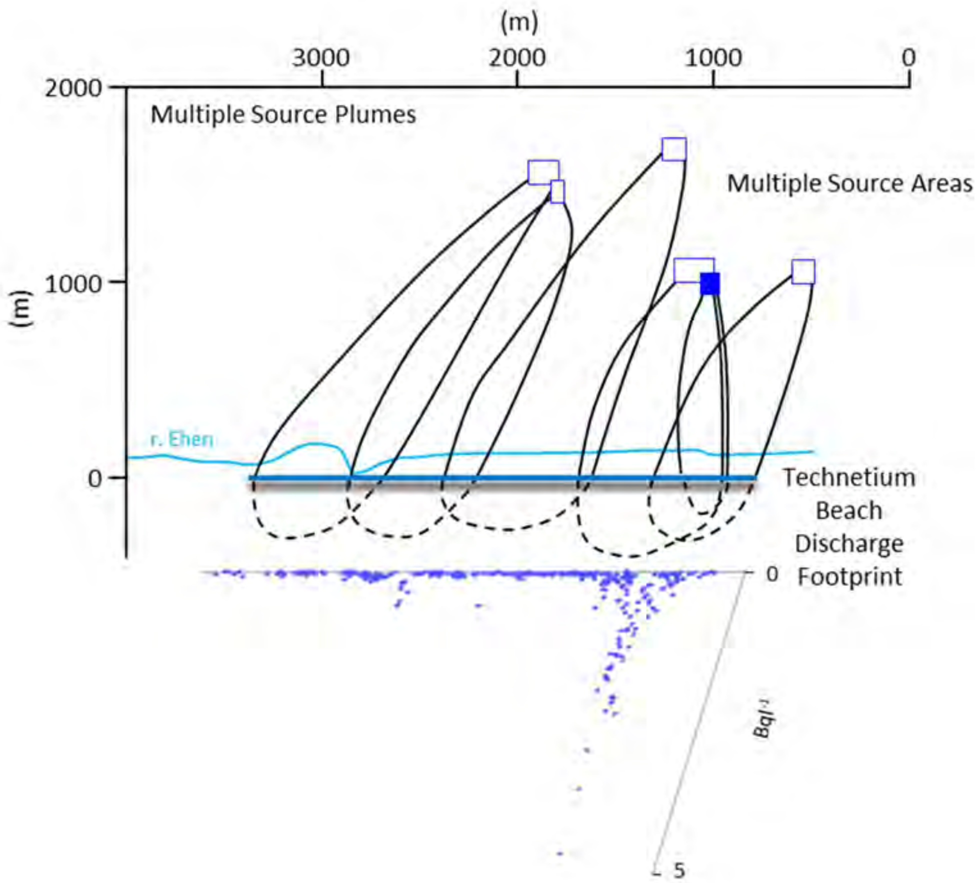

Figure 5-11 Interpretation of multiple technetium-99 source plume expression at the beach spring discharge area.

The conclusion of this simple transport modelling is that multiple sources of tritium are migrating in narrow plumes towards the coast and an expression of the different source signatures is seen at the beach spring discharge zone. Whilst the one-dimensional modelling is quick and a simple approach to solving the contaminant transport equation, this method is limited particularly as it does not take into account the spatial distribution of the tritium plume but more importantly doesn't allow us to refine the complexities and origin of the components of the beach spring waters exiting at the foreshore.

5.5 Tritium Migration Pathway Characteristics

5.5.1 Tritium in groundwater at the Sellafield site

The transport of tritium from source area towards the coast at Sellafield is complex since it involves numerous influences such as but not limited to; radioactive decay, dispersion, advection, changes in precipitation (recharge rates), and variable flow pathways in heterogeneous material of varying properties. Evidence found in the field studies allows the assumption that the tritium is migrating from multiple source areas in the central part of the site into the groundwater as tritiated water, travelling down-gradient towards the coast. The conservative nature of tritium means it is likely that it travels both in the shallow upper drift deposits, into the buried channel and exits in the area around the River Ehen and foreshore area. It is also likely to moves deeper in the underlying sandstone (lower aquifer) and eventually emerges as sub-marine discharges several hundred metres offshore.

Tritium time-series data collated from 1975-2018 from known Sellafield databases from site groundwater monitoring well sampling are presented in Figure 5-12 and shows an ongoing presence of the radionuclide in the groundwater under the Sellafield site, and provides confirmation that tritium is still migrating towards the coast. Interestingly there are high tritium readings across the 40 years span of data, yet the beach springs are showing stable low concentrations of tritium from the late 1990s onwards. This further supports earlier conclusions that the capping applied to the site has reduced the infiltration to a minimum and the plume has stabilised, possibly by also reducing the travel times and allowing radioactive decay coupled with natural attenuation controls to dominate more. The monitoring network has grown since 1975 and sampling techniques coupled with improved laboratory limits of detection have also improved the accuracy and hence more sample points are available and is indicated by the density of data increasing over the time-series.

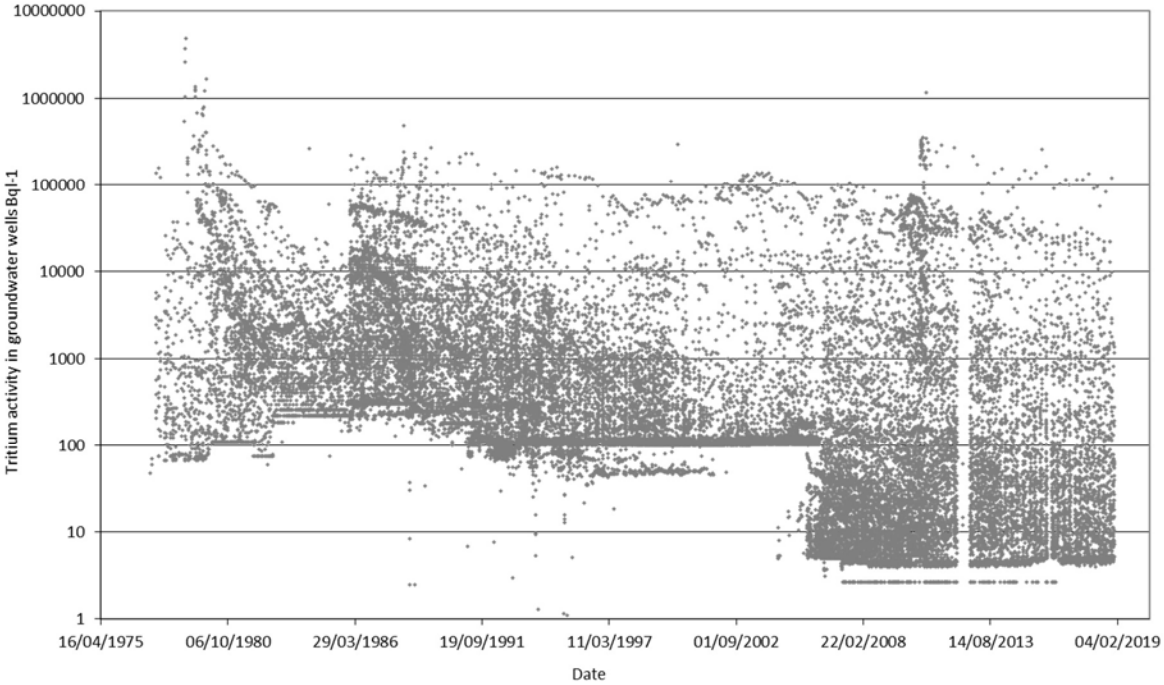

Figure 5-12 Tritium time-series data from the Sellafield groundwater monitoring well network (1975-2018).

We can further explore the pathway of the tritium by considering the site in transects between the source zone and the beach. Time-series tritium data in groundwater wells are analysed in transects perpendicular to the regional south-westerly groundwater flow in order to predict any future peaks of tritium that may surface in the beach springs. The estimated area of the primary source is shown by the red line in Figure 5-13, as well as the ‘transect’ locations and associated groundwater monitoring wells. There are some newer monitoring wells the ‘10,000’ series (post 2010) that have been drilled but they are excluded from the analysis and monitoring wells with longer temporal datasets were selected to enable comparison with the beach springs time-series data.

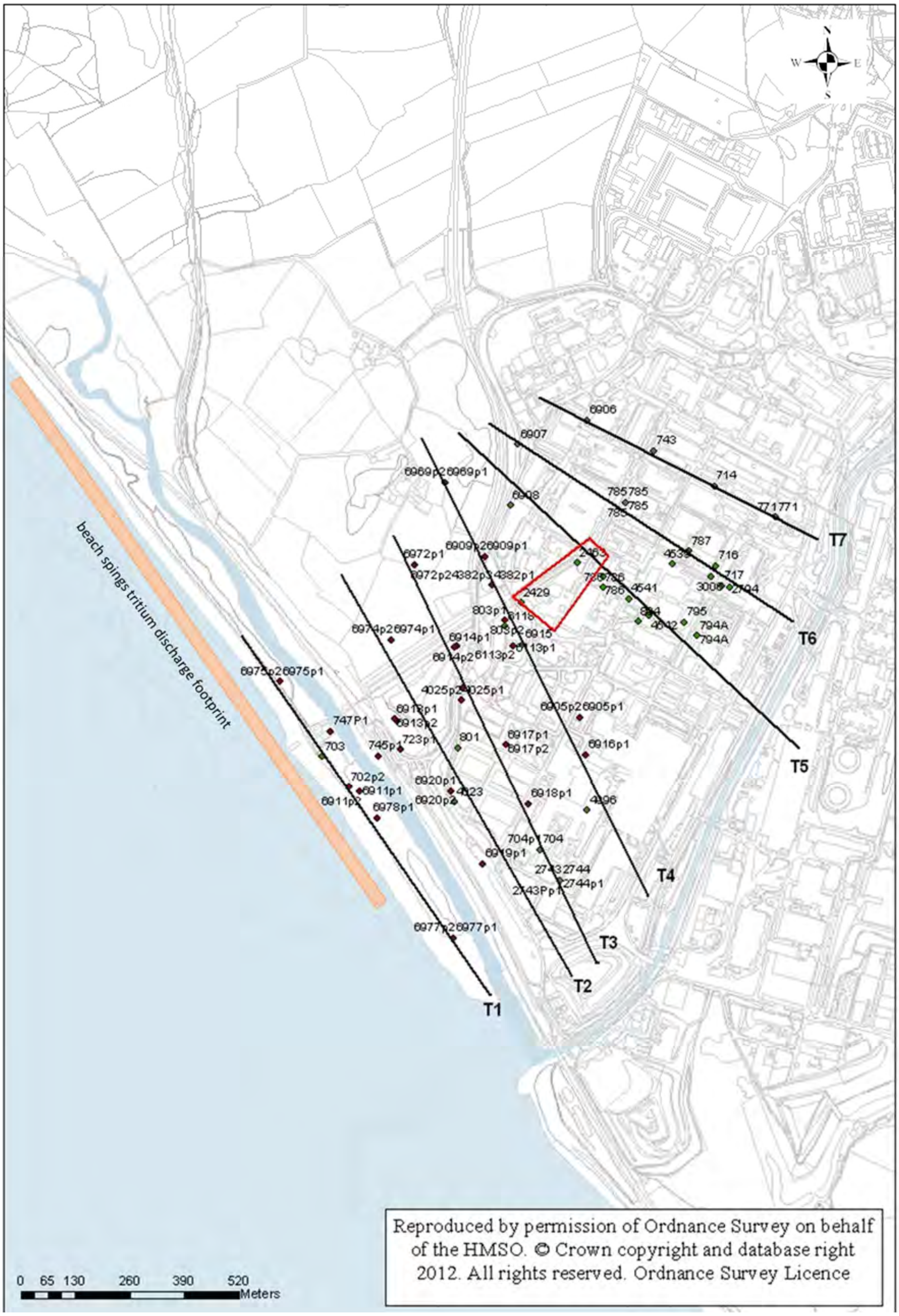

Figure 5-13 Selected transects for time-series tritium analysis in groundwater monitoring wells (with long temporal records), estimated area of primary tritium source areas marked in red.

Figure 5-14 shows the results of the monitoring wells that had tritium concentrations above the limit of detection. The peaks of concentrations are seen around transect 4 that is located down gradient of to the south-west of the contaminated zone (shown earlier in Figure 2-14). The monitoring wells across the individual transects do not show consistent levels of tritium activity and supports the conclusions about multiple narrow plumes. It is concluded that the tritium plumes are not yet in decline and continue to migrate towards the beach springs discharge zone.

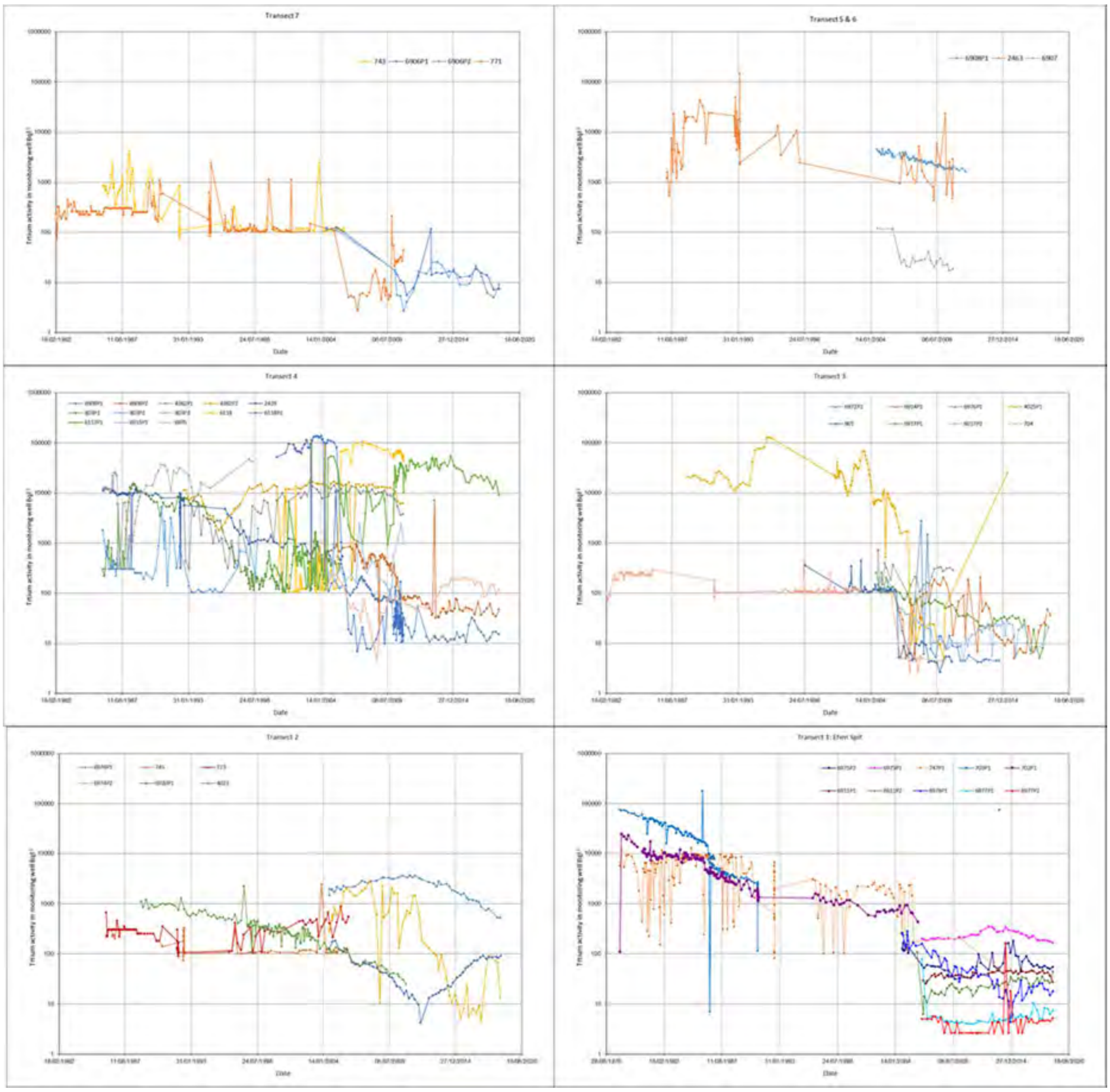

Figure 5-14 Tritium time-series in monitoring wells along transects from north of the site source area (transect 7) to the Ehen Spit monitoring wells (transect 1).

5.5.2 Modelling tritium pathways to the beach springs by inverse particle tracking.

Sellafield Ltd has a 'state-of-the-art' groundwater model (Connect flow) and integrated contaminant transport solution (GoldSim) calibrated with decades of contaminated land, geological, hydrogeological and groundwater data (Serco, 2010). The beach spring tritium sample data locations were used to represent a particle 'exit' point on the beach, by taking the centre point of the clusters of springs (Figure 5-15) and represent the spatial extent of the discharge footprint with the intention to inverse particle track groundwater pathlines from the beach springs back in towards site and predict the travel times to multiple source areas.

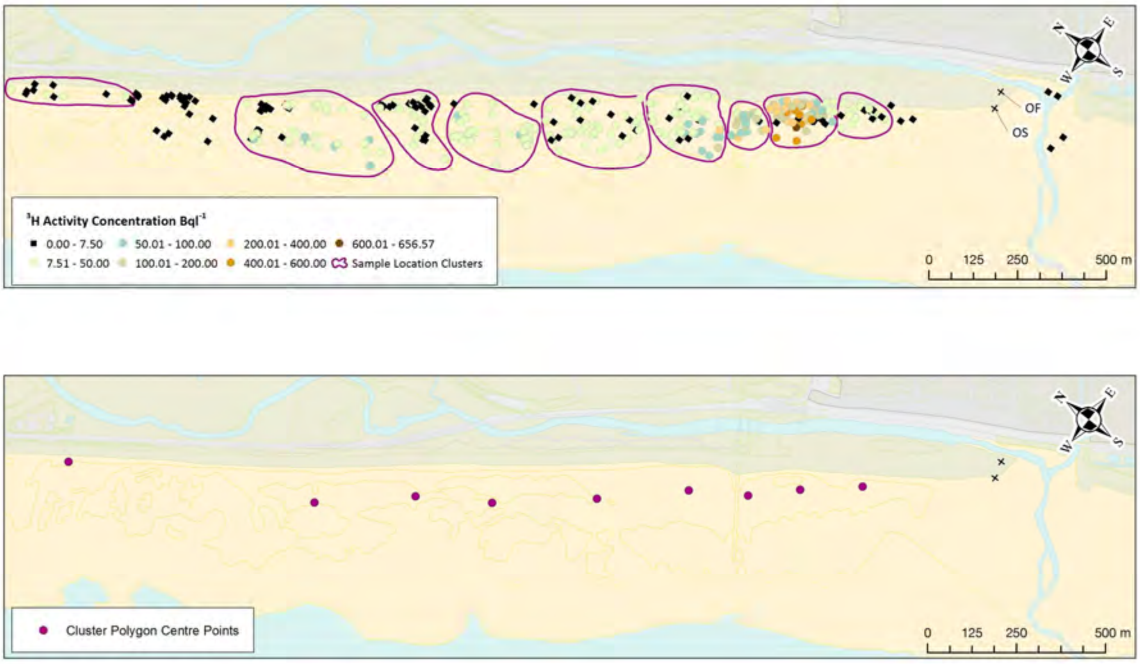

Figure 5-15 Approach to cluster samples point to single input particle exit point to represent the discharge footprint into the Sellafield groundwater model (cluster points 1-9 from N to S).

The model set up input parameters were based on the calibrated baseline groundwater flow and transport model for Sellafield site (Serco, 2010). The results of the model run are shown in Figure 5-16 and a very interesting key finding of this research study is that the beach tritium discharge footprint extends further up the beach than first thought and as such the northerly part of the footprint is outside the reaches of the Sellafield groundwater model flow field, which indicates that there may be a more complex groundwater flow regime in the north-west region of the site and not accounted for in the current conceptual model. The concurrent technetium-99 discharge footprint further supports this conclusion. A detailed investigation is recommended to examine the groundwater heads in that region again, as well as consider other geological controls such as faulting. The model estimated that the travel times from the source areas to the peak discharge zone 'A' range between 7 and 14 years (points 6-9) on Figure 5-16, and the upper range of 14 years is in reasonable agreement to the output of the analytical transport modelling in section 5.3.4 given the 3D Sellafield groundwater model is able apply better refinement of the system.

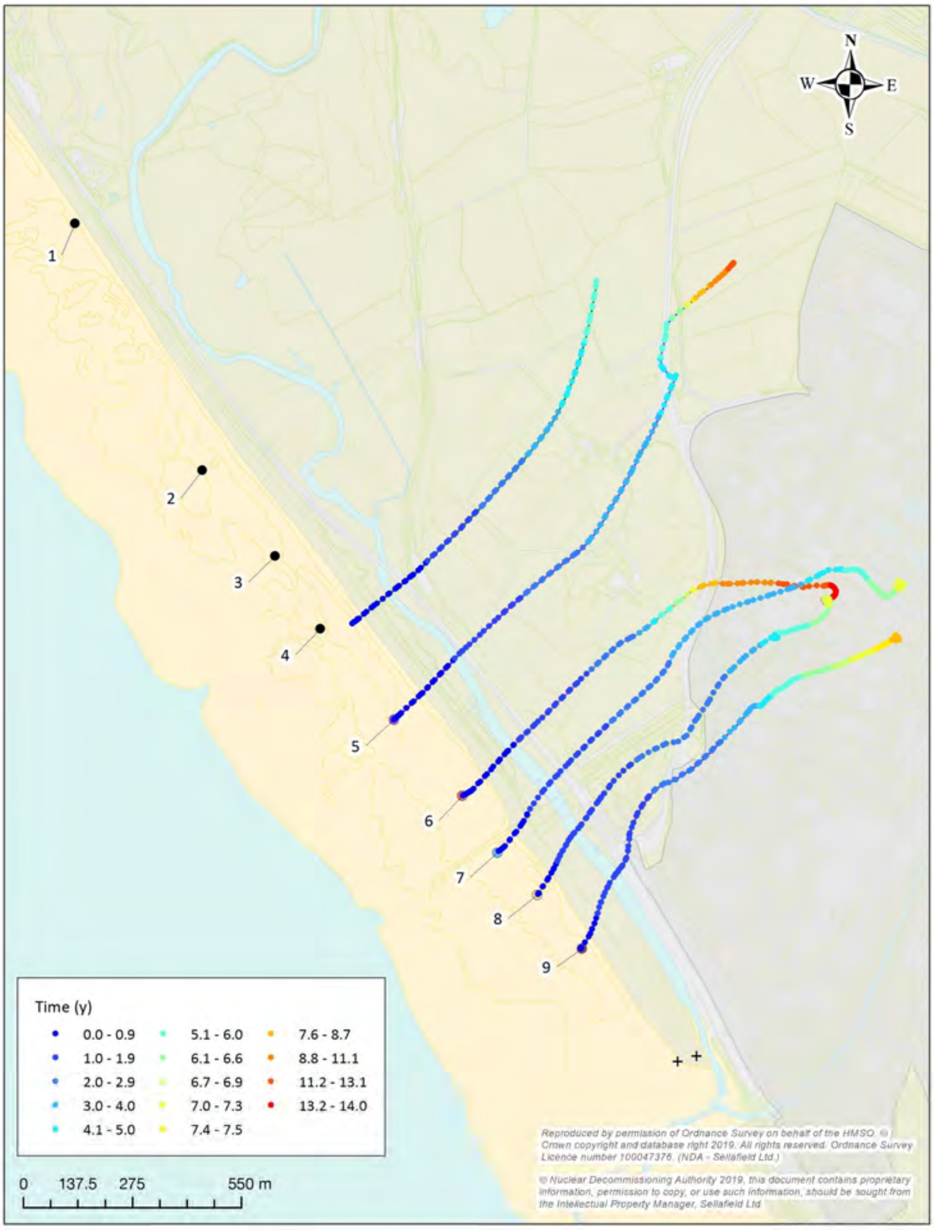

Figure 5-16 Model results for inverse particle tracking pathlines to inform travel times from tritium sources to beach spring locations.

5.6 Conceptual Model of Tritium Surfacing at the Beach Springs near the Sellafield Site

5.6.1 Synopsis of studies to inform the conceptual model

The previous chapters have summarised a range of studies to investigate the tritium arisings at the beach near the Sellafield site. The coastal interface dynamics near the Sellafield site remain a complex and difficult dual receptor area to interpret. There has been the added challenge of access to very large and exciting historic datasets but bounding scope direction to provide the right balance of interpretation and conceptualisation has provided difficult. Many new and interesting research questions have surfaced for the future research consideration. The utility of the inexpensive beach spring monitoring approach has provided new insights into the conceptualisation of the tritium and technetium-99 plumes travelling towards the coastal area. There is little risk to the public due to the low level of contaminant activities however system understanding is important to continually renew and update the groundwater contaminant risk assessments at the site. Table 5-3 provides a short synopsis of the studies and is presented as a set of claims with signposts to the supporting evidence.

Table 5-3 Synopsis of studies to inform the conceptual model development

	Claim	Evidence
Source	^{3}H and ^{99}Tc sources exist at the Sellafield site	• Review of archive information concluded several sources of differing magnitude with the primary source as the trenches for 3H and the sludge tanks for 99Tc • Groundwater monitoring data shows highest concentrations nearest the identified source areas in (Serco, 2010)
Pathway	Narrow contaminated groundwater plumes are migrating and merging towards the coast	• Transects show variable high levels of 3H still on the site • 1D transport modelling favours narrow plume hypothesis • Inverse particle back tracking shows pathlines to multiple source areas
Receptor	^{3}H and ^{99}Tc are emerging in the river and sediments of the River Ehen	• Liquid and sediment samples both show the presence of 3H & 99Tc • River bed tritium survey in (Whittaker 1977)
Receptor	^{3}H and ^{99}Tc are surfacing on the beach face as a seepage front, depicting a source	• Four decades of beach spring monitoring for ^{3}H has delineated a clear asymmetric discharge plume at the

	plume wider along the foreshore than predicted by the groundwater conceptual model	beach, supported by 10 years of ^{99}Tc data with a similar discharge footprint • 1D contaminant transport modelling using plume width characteristics favours a narrow source and plume • Long term temporal profile of tritium at beach spring from archived reports

5.6.2 Conceptualisation of tritium arrival at the beach springs

Building on the preliminary conceptual model presented in Chapter 2 and consolidating the technical conclusions of each chapter further refinements to the overall conceptual model is provided in Figure 5-17. The studies have supported the outcomes and conclusions made in the earlier investigations in the 1970-1980s and further improved the understanding of the spatial and temporal tritium discharge footprint at the beach springs, and supports the beach groundwater dynamics work carried out by Horn (2012, 2016) by tritium acting as a perfect tracer of the interactions between groundwater and beach face seepage zone when the water table decouples from the tide, described in more detail in Chapter 3.

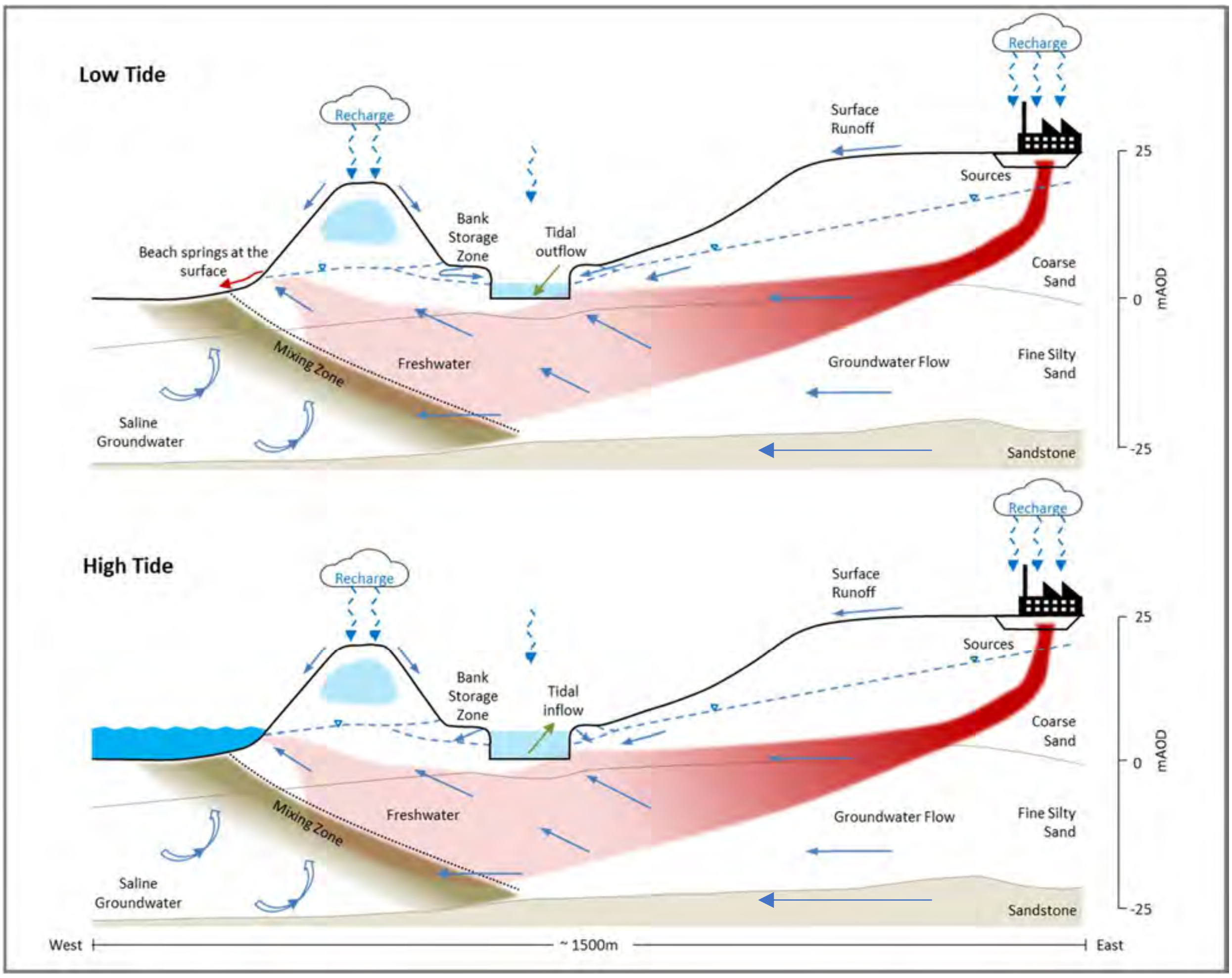

Figure 5-17 Conceptual model sketch of tritium plume arrival at the beach springs near the Sellafield site.

5.7 Conclusions

1) The spatial footprint and 1D contaminant transport modelling of tritium arrival at beach springs would suggest multiple plumes overlapping along the beach spring discharge zone, although the results were overly simplified and limited to be calibrated against the peak beach discharge zone.

2) The Sellafield groundwater model provided further insights by using the tritium discharge footprint to inverse particle back track the groundwater pathlines and lead to a key conclusion that the current Sellafield conceptual model does not account for a tritium plume stretching

up to 2600 metres north of the site and hence the groundwater flow regime and geological controls of the north-west region of site warrants further investigation.

3) Travel times between the source areas and the beach springs broadly aligned and in the range of 7 to 14 years.
4) The beach springs data does indicate that the tritium plumes are not yet in decline, all be it stabilised.
5) Further studies should be carried out to understand whether technetium-99 has also passed peak concentrations at the beach, or whether there is an increasing future trend, this is hard to ascertain due to shorter temporal records.

The key finding of this research study is that far-field geospatial analysis at the beach spring discharge zone has provided new insights into the source characteristics and groundwater flow pathlines from the Sellafield site. Many decades of near source and mid-field groundwater monitoring has developed and calibrated the Sellafield conceptual and groundwater flow model yet this has led to bias towards locations of groundwater monitoring wells located near to or on the site which has meant that the spatial extent of the tritium plumes has until this research study gone undetected and therefore demonstrates the utility of inexpensive far-field beach spring monitoring comparatively to the large investment required for the ongoing groundwater monitoring on the Sellafield site. It is recommended that nuclear sites located near surface or coastal waters adopt far-field seepage monitoring such as riverbed or beach spring monitoring to compliment the groundwater monitoring programmes.

5.8 Recommendations for Further Work

The priority area for further research is to understand and model the groundwater flow pathways to the north-west of the site to explain the extent of the beach springs discharge front northwards beyond the site. Emerging groundwater data from far-field of the nuclear site may also indicate a tritium presence and should be compared with the tritium beach discharge footprint. It is recommended that the Sellafield conceptual model is updated with the findings presented in this thesis.

CHAPTER 6 CONCLUSIONS AND FUTURE RESEARCH DIRECTION

This thesis set out to demonstrate the value of the diagnostic far-field beach spring monitoring and provide new insights into radionuclide contaminant source expressions at the coast and improve understanding of pathways to the beach receptor, to inform of future risks posed. The results show a discrete peak tritium discharge and, to one side, a lower concentration discharge spanning 2.5km along the beach foreshore. These accord with a potential known on-site former disposal source and the more dispersed impacts of multiple sources at greater distance and probable geological-hydrogeological-hydrological controls discerned. Whilst the tritium beach discharge record is long and potentially the earliest available of its type globally (starting in 1975), the rising limb and peak in time of beach tritium discharge was not captured; all data are on a declining limb of the breakthrough curve. It was hence not possible to fully constrain the minimum timeframes for transport of tritium from the main suspected source to the coast but were estimated to be 7-15 years in this study. From a regulatory perspective, the study has allowed beach spring monitoring at the site to be more effective by targeting measurements around the identified discharge area, and further investigation should continue. Tritium levels recorded in the recent years at the beach receptor are below the guideline risk level of 1.0 E-^{06}/year and within tolerable limits (HSE,2005). The data provides a good understanding of how the contaminant plume has interacted with the foreshore and provides strong evidence to the regulators that the contaminant source is abating. Chapter specific technical conclusions are made throughout the chapters, summary conclusions are made here:

6.1 Principle Findings and Conclusions

The key conclusions are presented against the objectives set out in Chapter one and are summarised below:

- *Investigate and collate evidence to propose a preliminary conceptual model (hypothesis) of tritiated groundwater surfacing at beach springs in the intertidal zone, near the Sellafield nuclear site*

A systematic review was undertaken to assess evidence for a tritium groundwater plume surfacing on a beach near the UK's oldest nuclear site. An initial conceptualisation of the evolution of a tritium plume related to multiple contributing source terms, by merging groundwater flow paths near the coastal intertidal zone (receptor) and surfacing in the intertidal zone in beach springs is presented based on all the information collated and interpreted during the review. Considerable material used in this review remains unpublished, and likely holds historical significance as one of the earliest coastal groundwater contaminant plume investigations in the UK, and possibly one of the earliest inland to be characterised along with a few landfill plumes in the 1980s.

- *Obtain new field data characterising the nature and significance of the tritium groundwater plume beach discharge footprint*

The lateral extent of the tritium and technetium-99 discharge footprints along the beach was further north of the Sellafield site than previously predicted by the groundwater monitoring network and the conceptual model, both of which suggest a more complicated groundwater flow regime in the north-westerly reaches of the site.

The key conclusion from this study is that the beach spring monitoring is acting to give an expression of the contaminant sources along the beach spring discharge zone. The beach spring monitoring also provides an inexpensive far-field monitoring tool at the receptor that should not be overlooked and a significantly lower cost than installation of groundwater monitoring wells. This could improve risk assessment of more attenuated migrating radionuclides.

- *Compare the observed modern discharge footprint to the collation of earlier archived beach spring discharge data spanning some four decades*

The discharge footprints for tritium and technetium-99 coincide spatially, and in reasonable agreement with the historic discharge data, although the modern field data shows a larger spatial footprint some 2600m in width. Technetium-99 is expected to be more retarded than tritium but appears to have dispersed laterally to the same extent as tritium and it can be concluded that technetium is as mobile as the tritium in the groundwater. Technetium-99 interactions with the river should be explored further due to the levels of concentrations reported.

- *Consider how the observed tritium discharge footprint informs future risks posed from more attenuated radionuclides to the coastal environment*

The results from the fieldwork presented in Chapters 3 and 4 provide evidence that relatively mobile radionuclide plumes from the Sellafield site will all migrate and reach the beach in spring water discharges and thus it can be concluded there is a hydrogeological connection between the source area and foreshore.

The lateral extent of the tritium and technetium-99 discharge footprint along the beach was further north of the Sellafield site than previously predicted by the groundwater monitoring network and the conceptual model, both of which suggest a more complicated groundwater flow regime in the north-westerly reaches of the site.

To date groundwater risk assessments deem it a low risk to the public and the findings of the fieldwork reported here do not change that conclusion, and the current groundwater plumes appear to have stabilised at steady state low concentrations, but it should be expected to see occasionally peaks at the discharge from the continuing presence of both tritium and technetium-99 in monitoring wells upstream of the beach. There is a future potential risk to the beach area should an unforeseen or accidental new leak happen at the site as mobile radionuclides will likely migrate along the same pathways as those from the historic leaks. Beach spring monitoring should be continued to monitor trends and the lateral extent of the plume.

- *Determine the contaminant sources, pathways, and breakthrough characteristics of the tritium groundwater plume at the coastal interface from the understanding developed by the far-field beach springs monitoring technique and refine the Conceptual Model and consider the wider utility of the approach*

The key finding of this research study is that far-field geospatial analysis at the beach spring discharge zone has provided new insights into the source characteristics and groundwater flow pathlines from the Sellafield site. Many decades of near source and mid-field groundwater monitoring has developed and calibrated the Sellafield conceptual and groundwater flow model yet this has led to bias towards locations of groundwater monitoring wells located near to or on the site which has meant that the spatial extent of the tritium plumes has until this research study gone undetected and therefore demonstrates the utility of inexpensive far-field beach spring monitoring comparatively to the large investment required for the ongoing groundwater monitoring on the Sellafield site. It is recommended that nuclear sites located near surface or coastal waters adopt far-field beach spring monitoring to compliment the groundwater monitoring programmes.

This study demonstrates the utility of beach spring monitoring of mobile radionuclide 'leader' plume discharge footprints as an indicator of future coastal risk. The approach is easy to implement and may effectively delineate the spatial expression of a plume discharge footprint in the coastal environment. The wider application is where evidence is known, or potentially imminent tritium discharges near coastal areas, that beach spring surveys are implemented. Where a long coastal foreshore can be monitored, it may usefully and easily provide a fingerprint of plume discharge that may constrain the geometry of the upgradient groundwater plume that is costly to investigate in high resolution by installation of groundwater monitoring wells. It may help to discern key source-pathway-receptor discrete linkages within the overall coastal discharge. Whilst there is significant value in securing historic foreshore tritium discharge data, a long data record may not exist at many sites and this study could be a useful reference case due to the indicative suite of legacy radionuclide contaminants in the sub-surface, by comparison found at most nuclear facilities worldwide. Historic temporal data from onsite boreholes more typically available should also be used to help at least partly constrain temporal trends.

Research herein suggests that contaminant plumes of mobile radionuclides should be in anticipated in the coastal zone and identification enables more accurate predictions of groundwater flow paths to potential receptors. Although previous studies have investigated migrating radionuclide groundwater plumes from nuclear sites as summarised in Table 2-2, to date no studies have elucidated relationships of radionuclide contaminant groundwater plumes with beach springs in coastal areas. Whilst the studies here presented in chapters 2-5 do not reflect every possible set of site conditions, it demonstrates that even without comprehensive system knowledge the samples from the beach springs monitoring are highly informative and led to new insights of contaminant transport at the Sellafield site which is arguably one of the most monitored in the world. This approach should be considered as part of a broader interdisciplinary spatial and temporal groundwater monitoring practises for improved system diagnostics for groundwater practitioners to understand vulnerability of coastal areas.

6.2 Recommendations for Future Research Direction

It is common when conducting fieldwork in remote locations over a protracted time frame that the study work will have a number of limitations due to the difficulties in resources, uncertainties in groundwater behaviours in an open hetergeneous system and conducting representative geochemical studies at the large field scale. Seasonal trends were particularly difficult to ascertain during this study even though it was conducted over 10 years, likely due to the periodicity of sampling. Further geochemical studies would also prove useful particularly in the river Ehen area, as well as offer a unique opportunity to study hyporheic interactions using radionuclide groundwater contaminants to trace flow interactions. Further research direction should be specific and targeted experimental fieldwork to further hydrogeological system understanding.

Detailed suggestions of limitations and further work are presented in each Chapter and it is recommended that the following specific studies are progressed in future as a matter of priority to support the ongoing protection of the Sellafield coastal area, and the new knowledge translated to the broader groundwater management application:

(1) The relationship between tritium and technetium-99 groundwater plumes and the river Ehen hyporheic zone should be investigated further by installation of piezometers and sampling carried out along the riverbed and the system understanding updated in the Sellafield conceptual model.

(2) A investigation of water levels to understand the flow regime at the groundwater-river-beach interface should be repeated and include river and groundwater level data loggers installed in and beyond the normal tidal limit, at a high temporal resolution.

(3) Higher resolution temporal monitoring of technetium-99 is required to understand the short term tidal and seasonal variation of the technetium-99 this will then allow the longer term trend to be assessed with more confidence. A longer temporal record is needed than the results of this study both at the beach springs and in the connecting pathways.

(4) The final priority area for further research is to understand and model the groundwater flow pathways to the north-west of the site to explain the extent of the beach springs discharge front northwards beyond the site. Emerging groundwater data from far-field of the nuclear site may also indicate a tritium presence and should be compared with the tritium beach discharge footprint. It is recommended that the Sellafield conceptual model is updated with the findings presented in this thesis.

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ELECTRONIC DATA APPENDICES

Appendix 1	Conference posters: A1.1: Determination of the risks posed by potential radiological contaminated groundwater discharges to coastal and river receptors: A strategy for investigation. A1.2: Reconstruction of a field-scale tritium groundwater plume at a UK coastal nuclear industrial site.
Appendix 2	A2.1: Systematic review spreadsheet for archive research study.
Appendix 3	A3.1: Sampling procedure & risk assessment for the beach springs monitoring study, and example chain of custody form A3.2: Sampling protocol/plan for the river-beach groundwater flow interactions study A3.3: Beach springs field measurement data and tritium analysis results A3.4: River Ehen sediment sample analysis results A3.5: Data logger processed data results for beach monitoring wells
Appendix 4	A4.1: Beach springs field sample data technetium-99 analysis results A4.2: River Ehen sediment & liquid sample technetium-99 analysis results