SEDIMENT SOURCE APPORTIONMENT UNDER DIFFERENT SPATIAL FRAMEWORKS IN AN AGRICULTURAL WATERSHED IN ATLANTIC CANADA

BY

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ABSTRACT

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Sediments negatively impact the quality of surface waters and are a significant source of contaminants, such as nutrients and pesticides in agricultural watersheds. Sediment fingerprinting is a relatively recent technique capable of determining the origin of suspended sediment. In this thesis, we investigated the sources of suspended sediments in a predominantly rural watershed in Atlantic Canada. Our first objective was to determine sediment source apportionment estimates by treating the watershed as a single catchment, and making the assumption that conditions affecting source production and transport, from the land to the stream, were uniform across the watershed with regards to geology, pedology, hydrology, weather, vegetation cover and land management. For the first objective, suspended sediments were collected at a single target location for sediment apportionment (main outlet) and used to represent sediment dynamics throughout the entire catchment. For the second objective, we examined not only the whole watershed but also sub-watersheds within it, in order to better understand processes affecting sediment dynamics and to determine if there was spatial variation in the origin of suspended sediments. The intent of our second objective was to explore different spatial framework (i.e., sampling design) options as well as explore what each option was able to tell us about sediment dynamics at a watershed scale.

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Table of Contents

List of Figures

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List of Tables

1. INTRODUCTION

1.1 Background

The goal of this thesis is to provide a means to quantify sediment source apportionment in an agricultural watershed within the Potato Belt region of New Brunswick in Atlantic Canada. Black Brook Watershed (BBW) has been selected for fluvial suspended sediment origin and sediment source quantification determination to examine the influence of intensive potato (*Solanum tuberosum* L.) production on sediment fluxes within the channel of Black Brook. This watershed has been cultivated for over 60 years (Rees et al., 2007). Agricultural production in this area has evolved from mixed farming for dairy and livestock (grain, forages and pasture landuses) to mixed farming coupled with some row crop production, to intensive potato row cropping within the last 50 years (Rees et al., 2007). In Canada, potato is recognized as the most important cash crop ($848 million in 2003), with New Brunswick producing 13 % of the annual potato production (Rees et al., 2007). In the 14.5 km^2 BBW, over 5.3 km^2 of the land is occupied by potatoes (Yang et al., 2009). The scarcity of land for rotations has led to intensive potato production with potato-grain and potato-potato-grain rotations (Rees et al., 1999).

Black Brook Watershed has been identified as being prone to some of the most severe water erosion in Canada (Chow et al., 1999). In this area, the terrain is undulating, with long and steep slopes, and these slopes are overlain by easily eroded soils (i.e., coarse to fine-textured dense, compact glacial till). Row cropping of potatoes in combination with a lack of soil conservation measures has been demonstrated to exacerbate water erosion, and result in a loss of high-quality topsoil (Coote, 1986). In BBW, degradation from intensive potato production on easily eroded soils has been estimated to cause soil losses of 17-40 t ha^{-1} yr^{-1} on intensively cropped potato fields (Chow et al., 1999). These soil losses may translate into crop yield reductions of more than 30 % (Chow, 1990). In the 1980s, the economic cost resulting from reduced crop yields were estimated to be $10-12 million dollars annually (Fox and Coote, 1986). Eroded soils assumed to be originating from these cultivated fields have also been linked to excessive contributions of sediments and nutrients into local surface waters. Sediment yield in BBW was estimated by Chow et al. (2011) at 181.6 t km^{-2} y^{-1} with a mean yearly total discharge of 0.587 million m^3 km^{-2} during periods from 2003 to 2007. Black Brook receives high quantities of sediment resulting in its streambed often being covered with a thick layer of fine sediment near harvest season. High flow rates in spring, however, flush the streambed clean, sending silt that was temporarily stored in Black Brook downstream into the St. John River and towards higher populated areas, with some of these areas depending on the St. John River as a source for drinking water.

Black Brook Watershed has been established as a national benchmark watershed to monitor and examine relationships between soils, stream discharge, sediment yields, chemical loading, topographical features and potato cropping practices due to the severe soil and water degradation within the watershed (Yang et al., 2009). The establishment of BBW as a benchmark watershed has stemmed many research studies since monitoring began in 1992 (Yang et al., 2009). The majority of studies have focused predominantly on upland erosion from agriculture in combination with the development of Beneficial Management Practices (BMPs) to protect surface waters from upland erosion. For example, according to Yang et al. (2009) it is estimated that approximately 50 % of cultivated lands and 24% of the total watershed area in BBW was covered by flow diversion terraces as a BMP by 2005. Flow diversion terraces are designed to combat against upland erosion from agriculture, by preventing rills from developing into gullies, reducing sediment delivery by deposition, and trapping much of the sediment eroded from areas between the terraces by reducing long slopes into shorter segments (Yang et al., 2009). However, to the best of our knowledge, none of the studies conducted in BBW have confirmed that the majority of sediments are indeed originating from upland sources in order to verify the need for flow diversion terraces. It is possible that sediments may be originating, for example, from streambank sources which would, thereby, require the implementation of much different BMPs for erosion mitigation purposes. Understanding where the majority of sediments are originating from allows for more effective BMPs to be implemented; thereby increasing efficiency and reducing costs which are often of utmost concern for producers when deciding to implement BMPs.

Much of the data obtained to date on the efficiency of BMPs at protecting soil and surface water quality has been obtained by conducting field experiments (e.g., Chow et al., 1999) or using integrated models such as the Soil and Water Assessment Tool (SWAT) (e.g., Yang et al., 2009). Integrated models, such as SWAT, are frequently used to identify areas within a watershed contributing excessively high amounts of pollutants per unit area. The SWAT model can simulate complex processes of erosion, hydrology, and nutrient losses and is capable of simulating management practices, such as, fertilizer applications, tillage operations, crop rotations, making it a useful tool for the evaluation of BMPs. To ensure the long-term effectiveness of agricultural BMPs a good understanding of spatial and temporal variations in land management practices, soil characteristics, topography and climatic conditions is required in order to assess the impacts of BMPs on water quality; however at a watershed scale this is often difficult using the methods mentioned above (Yang et al., 2010). Therefore, there is a need to assess the effectiveness of BMPs by identifying and quantifying sources of suspended sediments throughout the watershed in order to manage sediments which are adversely impacting the quality of soil, surface water and terrestrial and aquatic environments more successfully.

1.2 Literature review

Impacts of excessive fine sediments (< 63 μm) on water quality and aquatic and terrestrial environments are one of the dominant concerns facing the agriculture industry today. Excessive delivery of sediment to surface waters results in increased turbidity, reduced light penetration and transport of sediment-bound nutrients to surface waters. Excessive transport of sediment-bound nutrients, such as phosphorus, causes eutrophication of streams, rivers and lakes. With no clearly defined entry points, this type of pollution is hard to attribute to specific locations. To protect and improve water quality requires the identification of sediment sources and transport mechanisms within catchments. The sediment fingerprinting technique has been utilized to provide information on the origin of sediments within watersheds for decades (Walling et al., 1999). Sediment fingerprinting techniques are based on the idea that sediments entering the water have a physical, biological and/or chemical signature that reflect their source, and therefore can be used diagnostically to identify their origins (Collins et al., 1997; Walling et al., 1999). Multivariate approaches are used in which a large number of fingerprinting properties are measured and statistical analysis is conducted to determine the optimum number of fingerprinting properties that best apportion sediment to various sources (e.g., Koiter et al., 2013b; Barthod et al., 2015).

To discriminate between several potential sources, a large number of fingerprinting properties are recommended to be incorporated into a composite signature in order to provide reliable estimates of the relative contribution of different sources (Walling, 2013). The use of a composite fingerprint by combining independent signatures that respond very differently to environmental controls will result in better discrimination between potential sources and will provide more accurate sediment source apportionment (Collins and Walling, 2002; Mukundan et al., 2012). Especially, when it comes to the use of spectral-reflectance signatures (i.e., colour) signatures that have been identified as an emerging technique with a rapid and inexpensive means of investigating sediment sources, previous research from Barthod et al. (2015) and Martínez-Carreras et al. (2010) recommend combining colour fingerprints with more conventional fingerprints (e.g., geochemical elements and fallout radionuclides), to improve sediment source apportionment. To the best of our knowledge there has been few sediment fingerprinting studies conducted to date, with the exception of a study conducted by Krein et al. (2003) which combined colour fingerprints with fractal dimensions, which have utilized colour fingerprints with conventional fingerprints. In the current study a composite fingerprint which included radioisotope Cs-137, colour, and particle size and shape were utilized. The individual signatures making up the composite fingerprint were chosen because of their variation from one another with regards to environmental controls.

Caesium-137 is a fallout radionuclide which was deposited atmospherically during either nuclear weapon testing or nuclear accidents (Zapata, 2002). Cs-137 was deposited relatively evenly at a regional scale. Due to its surficial deposition and limited mobility, it makes an excellent tracer in terms of discriminating between surface and subsurface soils. It is absorbed to clay and organic particles in the topsoil where it acts as a tracer for soil movement (Devereux et al., 2010). The distribution of Cs-137 in undisturbed soil profiles shows an exponential decrease with soil depth, whereas in plowed soils it shows a uniform distribution through the plow layer (Ritchie et al., 1970). Therefore, Cs-137 is capable of providing discrimination between cultivated and non-cultivated land and between eroded and depositional areas as tillage and erosional processes redistribute the tracer across the landscape with physical processes being the main cause of Cs-137 redistribution.

The use of sediment colour as a fingerprint property to determine sources of sediments is an emerging technique that can provide a quick and inexpensive method to investigating sediment sources (Bathod et al., 2015). Research suggests that colour has the potential to be reasonably accurate in characterizing the properties of soil (Islam et al., 2003; Torrent and Barron, 1993). Colour is three-dimensional (Hunt and Pointer, 2011). According to Barthod et al. (2015), colour can be represented in colour space models, whereby individual colours are specified by points in these spaces (Barthod et al., 2015). Those parameters are continuous physical variables that can be used to describe soil colour and thus can be used to quantify the sources of sediment in the application of sediment fingerprinting (Barthod et al., 2015).

Particle size and shape was one of the earliest fingerprinting signatures developed to allocate sediment sources in watersheds (Grimshaw and Lewin, 1980). Particle size characteristics are extremely important because they influence the ability of sediments to absorb contaminates and nutrients (Liu, 2014). As a signature, particle size and shape can be used if greatly contrasting texture origins exist for sediments. These differences in texture can be used to trace sediments back to their sources (Davis and Fox, 2009).

A critical assumption underpinning sediment fingerprinting in watersheds is that differences in: geology, soil type, and/or land management practices imprints a signature on catchment soils that are decipherable from one source to another. It is assumed that the composition of sediment including their physical, biological and chemical properties, does not change (i.e., remains conservative) as the sediment moves through the landscape such that direct comparisons between sources and sediments can be made (Koiter et al., 2013a). Many sediment fingerprinting studies which assume signature conservativeness also assume that sediments collected at a watershed's main outlet, and conclusions based on these samples, may be extended to represent sediment dynamics throughout the entire catchment. However, the conservativeness associated with many fingerprinting signatures rarely occurs and sediment properties often change as sediments move through the landscape. High spatial and temporal variability often limits the ability to reliably link sediments back to their sources (Koiter et al., 2013a). Sediments may contain substantial spatial variability due to influences from climate, hydrological, topographical and geomorphological characteristics of the watershed (Koiter et al., 2013b). Furthermore, there can be a major disconnect in the sediment cascade between the headwaters and the outlet of a watershed (Koiter et al., 2013b). These complexities associated with sediments and their transport in a watershed need to be considered in both sampling strategies and the interpretation of data. There is, therefore, a need to explore the ability to utilize the fingerprinting technique to achieve a higher resolution in terms of spatial variations in sediment origin at a watershed scale. The use of a more conventional methods of sampling, where sediments are collected at the outlet alone, may provide insufficient information to support a meaningful conclusion on the sources of sediment for the entire watershed. Therefore, there is a need for further investigation of spatial framework options (i.e., sampling designs) for sediment fingerprinting studies taking place in large heterogeneous watersheds that are composed of several sub-watersheds.

1.3 Thesis objectives

The general objective of this study was to determine and quantify sediment sources in an agricultural watershed in Atlantic Canada by using the sediment fingerprinting technique. The first specific objective was to determine and quantify sources of suspended sediments using a more conventional sediment fingerprinting spatial framework, where suspended sediments are collected at the main outlet of a watershed and used to represent sediment dynamics throughout the entire watershed. The second objective was to examine several sub-watersheds within the BBW to examine the ability to utilize the fingerprinting technique to achieve a better understanding of processes affecting sediment dynamics and to determine whether a higher resolution in terms of spatial variations in sediment origin within the overall basin would provide additional information that was lacking by using the conventional sediment fingerprinting method. As part of the second objective, different spatial framework options were explored as well as what each option was able to demonstrate about sediment dynamics at a watershed scale.

1.4 Organization of thesis

Each objective mentioned is the focus of a separate chapter in this thesis and each chapter is written as a separate manuscript.

Chapter 2 focuses on the use of a conventional sediment fingerprinting spatial framework for sediment source apportionment, where suspended sediments are collected at a single target location for apportionment (i.e., the main outlet of BBW) and used to identify the main areas contributing to instream suspended sediments within the predominantly cultivated BBW located within Atlantic Canada.

In Chapter 3, several spatial frameworks were examined by considering several sub-watersheds within the greater BBW, to identify whether the sediment fingerprinting technique could achieve a higher resolution in terms of sediment apportionment within the overall basin in order to better understand processes affecting sediment dynamics at a watershed scale.

In Chapter 4, conclusions of this study and recommendations for the future work are discussed.

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2. USING SEDIMENT FINGERPRINTING TO IDENTIFY SOURCES OF SEDIMENTS IN AN AGRICULTURAL WATERSHED IN ATLANTIC CANADA

Abstract

Knowledge of sediment sources is required for sediment management practices and for our understanding of sediment fluxes within channels. Investigations have shown that a number of soil properties, including physical, biological, and chemical characteristics, may be used as "fingerprints" to trace sediments back to their source. Spectral-reflectance colour coefficients have recently been successfully used in fingerprinting studies. Colour-based fingerprints integrated with other physical fingerprints like particle size and shape and more conventional fingerprinting signatures, such as the radionuclide Cs-137, may offer substantial information on the origin of sediments.

The objective of this study was to determine the sources of suspended sediments in a mostly agricultural watershed in Atlantic Canada. In this agricultural watershed, it was unclear whether sources of suspended sediments were predominantly coming from upland erosion from agriculture or forest landuse or from streambank erosion.

Fine-grained suspended sediments were collected using an in-situ time-integrated suspended sediment sampler at the outlet of the 1,450 ha Black Brook Watershed in Atlantic Canada from 2008 to 2014. Results for sediment fingerprinting indicate that topsoil of cultivated land contributed the greatest amount of sediments (50-65 %), followed by streambanks within cultivated land (33-47 %), streambanks of forested land (1-2 %), and topsoil of forested land (0-2 %). However, the spatial framework employed to acquire these source apportionment estimates treated the watershed as one catchment and assumed uniform geology, pedology, hydrology,

weather, vegetation cover and land management. Due to these assumptions, suspended sediments that were collected solely at the main outlet of the watershed were used to represent sediment dynamics throughout the entire catchment. This spatial framework may incompletely represent the origin of sediments at a watershed scale. The results of this sediment fingerprinting study highlights the need for more robust spatial frameworks capable of representing sediment sources at a watershed scale more completely. The current study presents a need for a more robust spatial framework in order to capture spatial variations in the origin of sediments.

2.1 Introduction

Suspended sediments derived from agricultural land can adversely affect soil quality, surface water quality and aquatic ecosystems. Sediments may cause declines in crop yields, inputs of phosphorus and nitrogen into waterbodies and inputs of pesticides, leading to significant environmental issues (Russell et al., 2001). In the temperate climates of Atlantic Canada, many agricultural regions are characterized by rolling to undulating terrain, with long and continuous slopes, and these slopes are overlain by easily eroded soils (Su et al., 2011). In the Upper Saint John River Basin of northwestern New Brunswick, where the land supports intensive potato crop production, soil erosion has long been a major environmental issue.

The Black Brook Watershed (BBW) where over one-third of the land has been converted to potato crops, this land is prone to some of the most severe water erosion in Canada (Chow et al., 1999). Previous research by Chow et al. (1999) attributes the majority of this intensive soil erosion to the relatively shallow soils and the continuous potato planting on up-and-down slopes. Many past studies have shown significant erosion in cultivated fields including reports from Saini and Grant (1980), concluding that the average annual soil losses for continuous potato planting were estimated at approximately 17 t ha^{-1} yr^{-1} and reports from Chow et al. (1990) concluding soil erosion rates ranged from 1.2 to 24.3 t ha^{-1} yr^{-1}, therefore it has been assumed that instream suspended sediments are originating from upland erosion from cultivated fields. There has been discrepancies, however, between field level erosion measurements (e.g., Saini and Grant, 1980; Chow et al., 1990; Chow et al., 1999) and stream monitoring of sediment loading (e.g., Chow et al., 2011). Research by Chow et al. (2011) which compared suspended sediment loads in BBW to two other nearby watersheds including: 1) Little River watershed with forested land covering 77 % of the total watershed area and the remainder either agriculture (16.2

%) or residential/wetland (6.8 %), and 2) Upper Little River watershed with forested land covering 91.3 % of the total watershed area followed by wetlands (5 %) and agriculture (3.7 %). The study found that suspended sediment load increased with increasing agricultural intensity, with values of 181.6, 121.6, and 57.0 t km^{-2} y^{-1} for BBW, Little River watershed and Upper Little River watershed respectively. Many sediment fingerprinting studies (i.e., Koiter et al., 2013b; Gellis and Noe, 2013; Voli et al., 2013) have concluded that streambank sources were a significant source of suspended sediments in a variety of watersheds; therefore it is possible that streambank erosion may be contributing substantially to instream suspended sediments in addition to upland erosion thereby leading to the discrepancy between field level erosion measurements and stream monitoring of sediment loading.

The results of this research would be the first of its kind in Atlantic Canada and would help to enhance the understanding of sediment sources and dynamics in the intensively cultivated BBW. The management of suspended sediments through the development of Beneficial Management Practices (BMPs) to minimize impacts of landuse, particularly agriculture, on water quality requires knowledge of the sources of sediment. A direct approach to determine the origin of sediments can be achieved by using sediment fingerprinting. This method is based on the assumption that sediment sources can be discriminated based on a set of properties or fingerprints that are comparable between both sources and the resulting instream suspended sediments, allowing for the determination of relative source contributions (Collins and Walling, 2004). These fingerprints are measured in both source and sediment samples using various laboratory methods and statistical and mixing models are then employed to estimate the contributions from potential sources.

The fingerprinting method involves the identification of specific sources through the establishment of physical, chemical or biological signatures that distinctively characterize each source within a watershed (Gellis and Noe, 2013). Fluvial sediments exhibit a composite of these signatures that allow them to be traced back to their origins (Gellis and Noe, 2013). By comparing the signatures of the suspended sediment samples to the signatures of the source samples, the contribution of each potential source can be determined. Successfully utilized signatures include: spectral reflectance (i.e., colour) (e.g., Grimshaw and Lewin, 1980; Martinez-Carreras et al., 2010), radionuclides (e,g., Peart and Walling, 1986; Walling and Woodward, 1992; Olley et al., 1993), stable isotopes (e.g., Papanicolaou et al., 2003; Rhoton et al., 2008), trace elements (e.g., Devereux et al., 2010; Koiter et al., 2013b), mineralogy (e.g., Motha et al., 2003) and magnetic properties (e.g., Slattery et al., 2000). A signature needs to be both measurable and conservative to be suitable in tracing the origin of sediments. According to Haddadchi et al. (2013) signatures should be capable of: 1) distinguishing between sediments derived from different origins, 2) staying constant with time or varying in a predictable way, and 3) staying constant along the transport path or varying in a predictable way. The use of a composite fingerprint by combining independent fingerprints that respond very differently to environmental controls, such as radioisotope Cs-137, spectral-reflectance colour coefficients, and particle size and shape signatures will result in better discrimination between potential sources and will provide more accurate estimates of sediment source proportions (Mukundan et al., 2012). There has also been a need identified by Barthod et al. (2015) and Martínez-Carreras et al. (2010) that the recent success of sediment colour as a fingerprinting signature should be integrated in a composite fingerprinting framework with conventional fingerprints to improve source discrimination. The following will describe the fingerprint properties.

Caesium-137 is a radioisotope that is the product of nuclear weapons testing during the 1950s and 1960s (Loughran et al., 1995) and nuclear accidents (e.g., Chernobyl in Ukraine in 1986) with peaks in the early 1960s. Caesium-137 was initially injected into the stratosphere, where it was mixed and circulated globally before being deposited onto the Earth's surface (Playford et al., 1993; Cambray et al., 1989). Cs-137 was deposited relatively evenly at a regional scale. It was absorbed to clay and organic particles in the topsoil where it acts as a tracer for soil movement (Devereux et al., 2010). Cs-137 is monovalent and thus has a small hydrated radius and high ionic potential which allows adsorption on soils and sediments to be rapid and strong (Eyman and Kevern, 1975). Increased concentrations of competing ions such as Na, K and H slightly decrease Cs-137 adsorption potential (Ritchie and McHenry, 1990).

Cs-137 has been used as a tracer to provide independent information on erosion and sedimentation patterns and rates. Because few qualitative and quantitative techniques are capable of providing data on both erosion and sedimentation, Cs-137 has become a very popular technique for many erosion and sedimentation studies. The distribution of Cs-137 in undisturbed soil profiles exhibits an exponential decrease with soil depth whereas in plowed soils, a uniform distribution through the plow layer is observed (Ritchie et al., 1970). Therefore, Cs-137 is capable of providing discrimination between cultivated and non-cultivated land and between eroded and depositional areas as tillage and erosional processes redistribute the signature across the landscape with physical processes being the main cause of Cs-137 redistribution in soils and chemical or biological processes having minimal influences. In addition, Cs-137 is well-suited for use in heterogeneous watersheds, since its concentrations are effectively independent of soil type and underlying geology (Walling, 2005). The spatial distribution of Cs-137, in both vertical and horizontal planes across the landscape, can be used to show erosion patterns in fields, the redistribution of soils within fields, and soil loss from fields (Zapata, 2002).

Colour is one of the most important attributes to describe and differentiate soils; research suggests that colour has the potential to be reasonably accurate in characterizing the properties of soil (Islam et al., 2003; Torrent and Barron, 1993). Variation in soil colour is caused by differences in: organic matter, moisture conditions, chemical and biological weathering, and redox reactions (Udelhoven et al., 2003). Soil colour is affected by organic matter related decomposition producing black and brown compounds. Minerals, such as iron, are known to cause red, brown and yellow hue values, and manganese, sulfur and nitrogen can form black mineral deposits. Soil texture plays a significant role in soil colour due to its influence on organic matter.

One of the most important physical characteristics of sediment, which influences its ability to absorb contaminants and nutrients, is particle size and shape (Liu, 2014). Particle size and shape was one of the earliest fingerprinting signatures developed to allocate sediment sources in watersheds (Grimshaw and Lewin, 1980). As a signature, particle size and shape can be used if greatly contrasting texture origins exist for sediments. These differences in texture can be used to trace sediments back to their sources (Davis and Fox, 2009). Physical signatures such as particle size and shape offer many advantages to fingerprinting studies due to these properties being easily measurable and readily identifiable (Davis and Fox, 2009).

The objective of this study was to identify and quantify sources of suspended sediments in the mostly agricultural BBW using the sediment fingerprinting technique. By using a single target location for sediment source apportionment, the aim of the current study was to observe how suspended sediments captured at the main outlet of the watershed represented sediment dynamics within the BBW.

2.2 Materials and Methods

2.2.1 Study Area

Black Brook Watershed is located in northwestern New Brunswick, just north of Grand Falls, near the town of Saint-André (47°05' to 47°09' N and 67°43' to 67°48' W) (Mellerowicz et al., 1993).

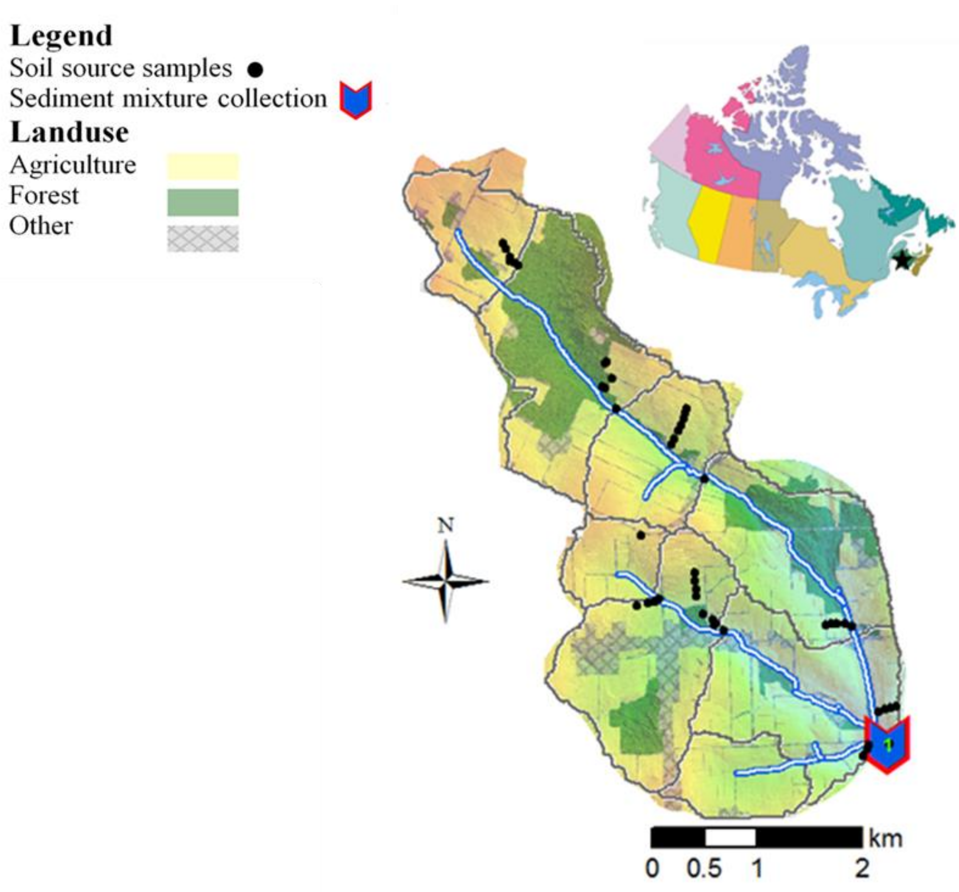

Figure 2.1 Land use and terrain of the Black Brook Watershed showing the location for sediment source samples and the location for the suspended sediment sampling

The BBW covers 1,450 ha and is approximately 7.5 km long and 3.5 km wide (DesRoches et al., 2014). Black Brook is a tributary of the Little River, which is located within the 380 km^2 Little River watershed. Black Brook Watershed and the greater Little River watershed discharge into the Saint John River. These watersheds are part of the Upper Saint John River Valley Ecoregion in the Atlantic Maritime Ecozone (Marshall et al., 1999). The climate is moderately cool boreal with a soil moisture regime ranging from humid to perhumid, with frost periods lasting for approximately 120 days (Mellerowicz et al., 1993). The mean annual temperature is 3.2 ºC (Environment Canada, 2012). The region has a mean annual precipitation of 1,134 mm with monthly averages ranging from a low of 64.6 mm in February, to a high of 111.6 mm in July (Su et al., 2011). Approximately one-quarter of precipitation in this region falls as snow (Su et al., 2011). Although monthly precipitation is relatively uniform from April to September, frequent summer storms tend to relocate large amounts of soil within the watershed. According to Chow et al. (2011), over 85 % of the sediment yield in BBW that occurs between March and April is typically eroded from the fields during the summer months and deposited in lower slope positions adjacent to the river. Cumulative erosivity from June to September was over 74 % of the yearly total indicating that during the growing season the majority of rainfall erosion occurs. Topography in BBW consists of valleys along Black Brook with neighbouring plateaus. The elevation ranges from 180 to 260 m above sea level (Mellerowicz et al., 1993). The lower portion of BBW is characterized by a more prominent rolling topography compared to the central and upper portion with slopes of 5-16 % in the lower portion, 4-9 % in the central portion and 1-6 % in the upper portion (Mellerowicz et al., 1993).

Potential sediment sources were identified based on the net result of the interaction between parent material, topography, hydrology, climate, weathering processes, amount and type of vegetation and anthropogenic activities in combination with the composition and physical structure of soils. This made it possible to discriminate between multiple sediment sources using natural soil properties. These watershed characteristics and processes may result in a large diversity of potential sediment sources in heterogeneous watersheds such as BBW. Therefore, the spatial framework utilized in BBW aimed to capture the diversity in potential sediment sources by ensuring sampling took into account 1) parent material, 2) soil types, and 3) landuse.

The advance and retreat of glaciers during the most recent Glacial Ice Age resulted in surface glaciofluvial deposits accompanied by morainal till deposits. This has characterized the western portion of Saint-André with rill wash composed of mixed sand, stones, reworked till and veins of gravel and silt and the eastern portion with underlain compact till with hummocks and ridges of stratified gravel, sand and silt (Mellerowicz et al., 1993).The basis of BBW is Ordovician and Silurian calcareous and argillaceous sedimentary rocks from the Matapedia Group (Rappol and Russell, 1989). Bedrock is weakly deformed showing evidence of very low grade metamorphism (Carrol, 2003). The base formations are dominated by thin-bedded, dark grey calcareous shale, siltstone, and fine grained sandstone, calciluite, minor to medium bedded calcareous sandstone and minor amounts of non-calcareous shale and siltstone deposited on a submarine slope environment (St Peter, 1978; Stringer and Pickerill, 1980). Because the signatures of potential source soils are largely related to parent material and their derived soil associations, BBW's specific geological features were taken into account in the sediment fingerprinting study.

Soil associations and their related soil type (subgroup level of the Canadian system of classification for the catena head on native soils) found in BBW are described in Table 2.1 and their general locations are depicted in Fig. 2.2. Black Brook Watershed is comprised of six mineral soils, including: Grand Falls (Orthic Humo-Ferric Podzol), Holmesville (Orthic Humo-

Ferric Podzol), Interval (Gleyed Regosols), Muniac (Orthic Humo-Ferric Podzol), Siegas (Gray Luvisol) and Undine (Mini Humo-Ferric Podzol), and one organic soil association, St. Quentin (Terric Mesisol or Terric Humisol). These soil associations vary substantially from one another with regards to mode of deposition, petrology, depth to compacted layer, texture, colour, drainage and coarse fragments (Table 2.1). Topsoils are poorly structured and dense compact subsoils are prevalent (Chow et al., 1999). Furthermore, extensive soil erosion has been observed in the Holmesville soil type (Mellerowicz et al., 1993). More information on the characteristics of the soil associations within BBW can be found in Mellerowicz et al. (1993) and soil types can be found in Rees et al. (2005).

Table 2.1 Soil in the Black Brook Watershed (adapted from Mellerowicz et al. (1993))

Soil Association	Soil type (subgroup level)	Mode of Deposition	Petrology	Depth to Compact Layer (cm)	Texture	Colour	Drainage	Coarse Fragments
Grand Falls	Orthic Humo-Ferric Podzol	Glaciofluvial	Noncalcareous Slate, Quartzite, Sandstone	>100	Gravelly Sand Loam	Olive Gray	Well-Poor	>50% gravels, channers
Holmesville	Orthic Humo-Ferric Podzol	Compact till	Sandstone, Quartzite, Argillite,Slate, Shale	20-70	Loam or Silt Loam	Yellow-Olive Brown	Well-Poor	15-30% cobbles, gravels
Interval	Gleyed Regosols	Alluvial	Unknown	>100	Silt Loam	Olive-Dark Yellow Brown	Very Poor	<5% gravels
Muniac	Orthic Humo-Ferric Podzol	Glaciofluvial	Calcareous, Slate. Shale, Quartzite, Sandstone	>100	Gravelly Sand Loam	Light Olive Brown	Well-Poor	>20% gravels
Siegas	Gray Luvisol	Compact till	Quartzite, Sandstone, Shale, Slate, Argillite	20-60	Loam or Clay Loam	Light Olive Yellow Brown	Well-Very Poor	<20% gravels
St. Quentin	Terric Mesisol or Terric Humisol	Organic	Forest Peat	40-160	Fibric Mesic	Dark Brown-Reddish Brown	Very Poor	Unknown
Undine	Mini Humo-Ferric Podzol	Residual (thin till)	Weakly Calcareous, Shale, Slate, Sandstone	65-100	Silt Loam or Loam	Light Olive-Yellow Brown	Well-Imperfectly	10-30% channers

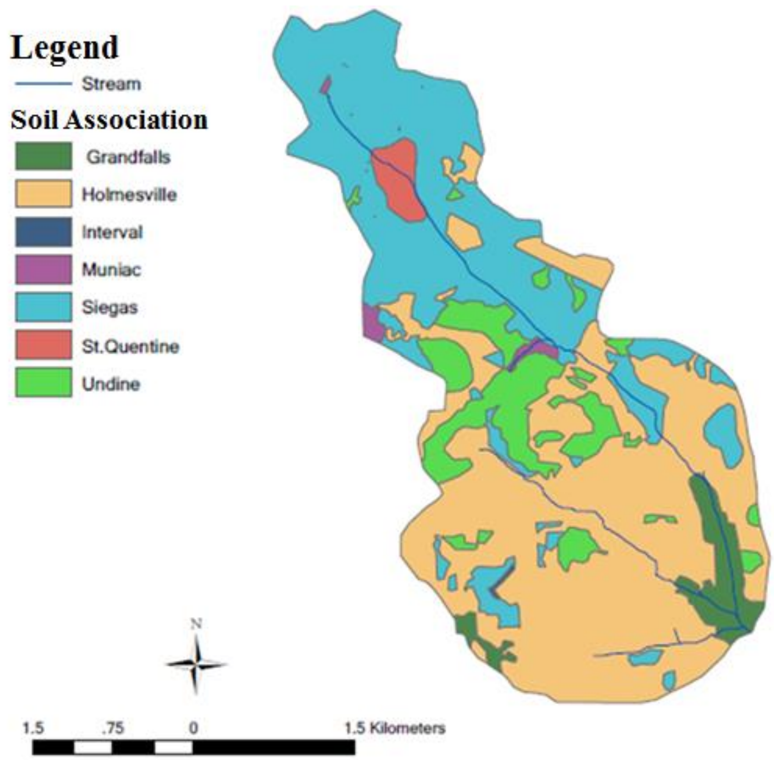

Figure 2.2 Main soil associations in BBW determined by Mellerowicz et al. (1993) using a scale of 1:10 000.

The landuse and boundaries for the eight sub-watersheds found in BBW are illustrated in Fig 2.1. Approximately 65 % (1,050 ha) of the land is used for agriculture, with potato grown in rotation with grains and hay for forage being the predominant crop (Chow et al., 2011). Approximately 21 % of the land is forested, with one major forest complex at the top of BBW while the rest is mainly riparian areas along the streams (Chow et al., 2011). The remaining 14 % of the land is comprised of residential areas, wetlands and/or roads (Chow et al., 2009).

2.2.2 Spatial Framework

A Simple Fingerprinting Study (SFS) was used to determine the relative contribution of sources contributing to suspended sediments within the mostly agricultural BBW. In the current study, a SFS refers to treating a watershed as a single catchment and assuming that conditions affecting production and transport for each source type are uniform across the watershed in terms of parent material, topography, pedology, hydrology, weather, vegetation cover and land management. The SFS used in BBW, is characterized by having one target location at the main outlet of the watershed which is identified as SW1 (i.e., sub-watershed 1) for the purpose of originating the sources of sediments. In many fingerprinting studies, (e.g., Poleto et al., 2009; Mukundan et al., 2010; Collins et al., 2012) sediments are collected solely at the main outlet of a watershed and conclusions based on these samples are used to represent sources contributing to the entire catchment. Multiple types of sediment (i.e., suspended and bedload) and/or multiple seasons of collection and/or multiple years of collection may be collected at the watershed's outlet to reach an adequate number of sediment samples. This sampling approach has become widely used among researchers because of its relatively quick and cheap identification and quantification of sediment source apportionment.

Source materials were collected in sampling areas located throughout the catchment of interest, typically upstream of the location intended for sediment source apportionment (Fig. 2.3).

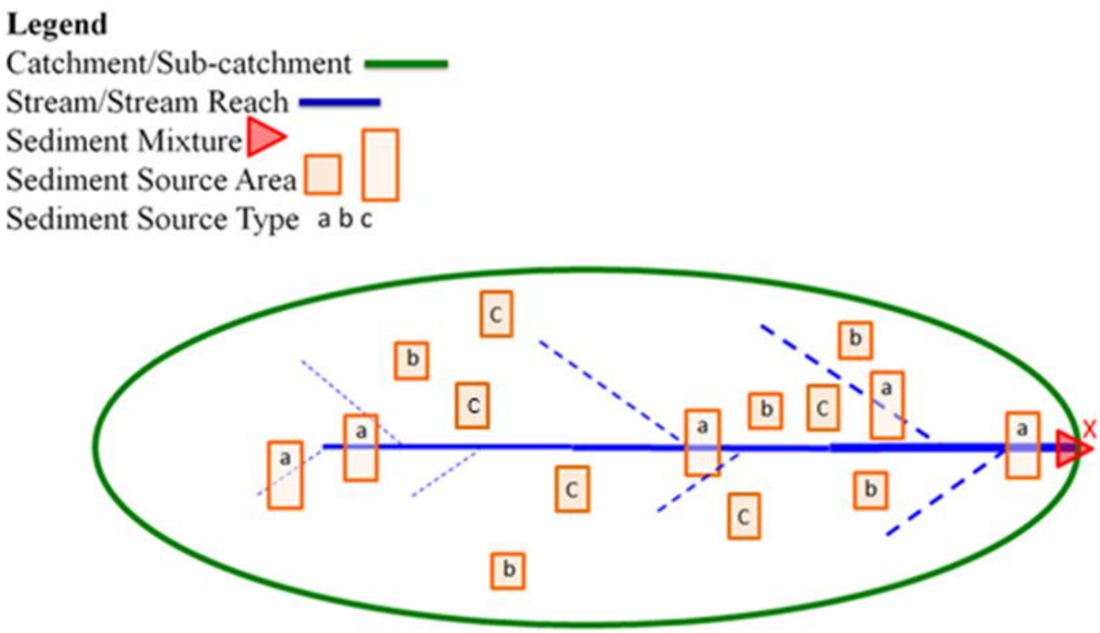

Figure 2.3 Single Watershed Source Sampling for a Simple Fingerprinting Study containing a single location of interest (X) for sediment source apportionment and three source types (a, b, c), each identified within five separate sediment source areas.

An example of how Single Watershed Source Sampling is utilized for a SFS containing one targeted location for sediment apportionment is illustrated in Fig. 2.3. Suspended sediments are collected at a watershed's main outlet (X) and all potential source materials are collected throughout the catchment in accessible areas with visible signs of erosion and high connectivity to the channel of interest. Three potential source types (a, b, c) have been identified as contributing to suspended sediments collected at the main outlet (X). A total of five sediment source areas were selected for each source type.

2.2.3 Collection of source materials and sediments

In order to evaluate the impacts of landuse on soil erosion and its contribution to fluvial suspended sediments, four potential sources of sediments were identified. These potential sources included: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambank, and 4) forest streambank. Source sampling locations were selected based on the following criteria: 1) visible evidence of erosion or soil degradation, 2) distance and potential connectivity to stream, and 3) access and safety (Koiter et al., 2013b). All potential source samples were collected upstream of SW1 (the main outlet), the location where suspended sediment samples were collected.

Soil samples were collected from the top 0-5 cm of the soil (i.e., topsoil A-horizon) to represent potential sediment source material. Soil samples were collected in transects, extended from the top of streambanks through the riparian zone and into nearby fields. Each transect was sampled along the toposequence parallel to the greatest slope gradient and consisted of 4-8 individual soil samples, depending on the extent of the erodible area. A total of nine transects were sampled within BBW amounting to a total of n=14 forest and n=36 cultivated soil samples. A corer was used to collect the majority of soil samples, however in places with many stones, shovels were used. In agricultural fields, the topsoil was sampled to the same depth as the tillage layer due to the soil being regularly plowed during the potato growing season. Therefore, the depth of the tillage layer (i.e., 40 - 50 cm) represents the potential erodible soil, requiring the entire depth to be sampled to acquire a representative source sample.

Streambank samples were collected near the outlet of several sub-watersheds within BBW (Fig. 2.1). Streambank samples were collected to construct a full streambank profile and consisted of multiple samples collected at the beginning at the top of the streambank working downward towards the base in 10 cm increments. Three streambank profiles were collected in five sub-watershed, totaling 15 streambank profiles for a total of n=67 sediment source samples for streambanks located in cultivated areas (i.e., cultivated streambank) and n=9 source samples for streambanks located in forested areas (i.e., forest streambanks). Streambank samples were collected in the autumn of 2014 when the stream flow was low. Samples were collected using a

10 cm by 10 cm box-core sampler. A scraper was used to make a smooth profile, free of vegetation, in order for the box-core sampler to be hammered into the side of the streambank perpendicularly. It was ensured that any soil which may had fallen from above was removed before the next sample was collected in order to reduce cross contamination. The intent of taking detailed streambank profiles was to ensure thorough representation of each streambank as a source of sediment in order to aid in the statistical and process-based interpretation of signatures (Koiter et al., 2013b).

Suspended sediments were collected seasonally, excluding winter, from 2008 to 2014. The collection period for spring occurred from April to mid-June, summer occurred from mid-June to mid-September and autumn occurred from mid-September to early-November. Samplers were designed after the Phillips et al. (2000) in-situ time-integrated samplers (Fig. 2.4), and were installed at SW1. The samplers were made of polyvinylchloride pipe with sealed caps at each end that contained 3 mm diameter inlet and outlet tubes. A funnel was placed over the inlet tube facing the direction of flow in order to streamline the sampler and reduce disturbance from ambient flow (Russell et al., 2000). A time-integrated sample was collected as water and suspended sediment entered the inlet tube and moved toward the larger polyvinylchloride pipe where the reduction in flow velocity induced by the change in cross-sectional area caused a reduction in flow velocity, thereby, causing sedimentation and the collection of the time-integrated sample. Samplers were attached to concrete blocks and chained to nearby trees to anchor samplers to streambeds and reduce the likelihood of sampler destruction or loss. Sediment samplers were inspected every two weeks for damage and tube blockages.

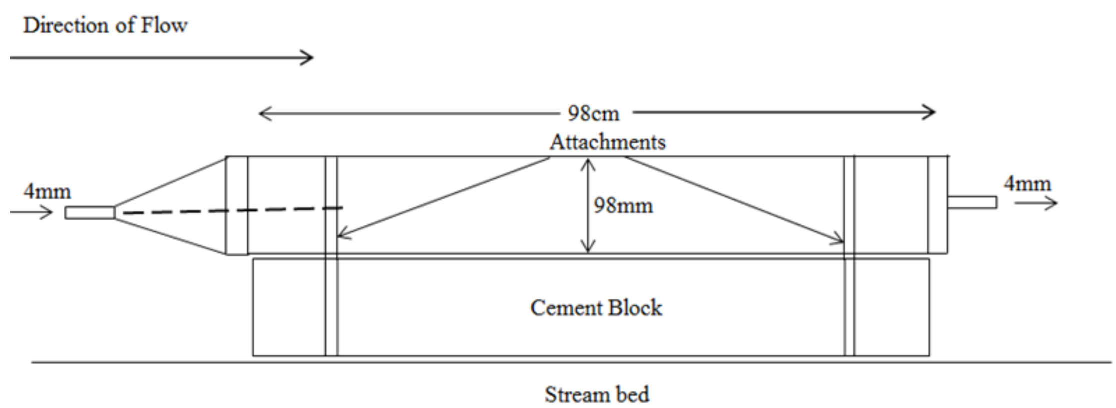

Figure 2.4 Cross-section of an in-situ time-integrated suspended sediment sampler used at SW1 in BBW, adapted from Phillips et al. (2000).

Sediment samplers were installed in sections of the stream deep enough to completely submerge the samplers in order to reduce the likelihood of air bubbles and to maximize suspended sediment collection. Samplers were typically installed within the midsection of a channel unless factors such as safety or high flow rates restricted access.

2.2.4 Laboratory analysis

Suspended sediments and source materials were analyzed for a variety of fingerprints including radiochemistry (Cs-137), spectral reflectance (colour) and particle size and shape parameters. Prior to analyses, all source materials were air-dried and sieved to < 2 mm to remove coarse fragments. Samples destined for spectral reflectance analysis were further sieved to < 63 µm. Wet samples were dried at 30 °C for upwards of 3 days. After samples were dried, they were stored in plastic containers until further analyses. Suspended sediment samples were initially emptied into buckets and left to settle for over 7 days in accordance to Stokes' Law to ensure that fine particles were given adequate time to settle. Following settling, water was carefully siphoned off, leaving a small amount of sediment and water. This slurry was then transferred to a pan for drying and subsequent analysis.

2.2.4.1 Radiochemistry and gamma spectroscopy

Caesium-137 activity was measured using gamma spectrometry with high purity germanium gamma spectrometers (Canberra) located at the Fredericton Research and Development Centre, Agriculture and Agri-Food Canada in New Brunswick. The activity was measured based on the germanium receiving a signal (662 keV) within the detector from the gamma photons emitted by radionuclide Cs-137. The crystal located within the detector sends a signal to the multi-channel analyzer system, where the emission counts were plotted against the energy of Cs-137. The activity of Cs-137, the number of nuclear disintegrations per second, was measured in Bacquerels (Bq kg^{1})

The suspended sediment samples were analyzed on one of two germanium well detectors, while the soil and streambank samples were run on a broad energy germanium detector. Suspended sediment samples were analyzed in plastic tubes at a height of 2 cm, and soil samples were analyzed in plastic cups at heights ranging from 1-8 cm depending on the amount of soil available. The weight, height and geometry of each sample were recorded and entered into the software before each analysis. The majority of the samples were counted for approximately 24 hours, while some low mass samples were counted for 48 hours. Cs-137 concentrations were decay-corrected, with all source and sediment samples corrected to January 1, 2010. Prior to sample runs, each detector was calibrated using reference standards supplied by the International Atomic Energy Agency (IAEA) in order to test the analytical performance of each detector and to produce the necessary variables required for inter-laboratory comparison.

2.2.4.2 Spectral-reflectance and spectroradiometery

To obtain colour properties, spectral readings were taken over a 350-2500 nm wavelength range using a spectroradiometer, ASD FieldSpec Pro (ASD FieldSpec Pro, Analytical Spectral

Device Inc.). A 10 cm diameter transparent plastic support was used to hold the sample that was smoothed with a straight-edge in order to reduce shading. The sample completely covered the plastic support to ensure that the colour coefficients obtained represented the sample and not the reflection from the transparent plastic support. A white reference, known as the Spectralon standard, contained certified reflectivity and was used to calibrate the spectroradiometer prior to each sample run. The sample and Spectralon standard were lit with a white light source (1000 W quartz halogen lamp [12 VDC, 20 Watt] mounted on a tripod at a distance of 10 cm) (Barthod et al., 2015). A fiberoptic cable was mounted 2 cm away and at an angle of 45° from the Spectralon standard followed by the sample (Barthod et al., 2015).

The absolute bidirectional reflectance spectra were obtained by using RS3 software by multiplying the raw reflectance spectra by the certified reflectivity of the Spectralon standard. Ten absolute reflectance spectra were collected for each sample and an average was computed. The Commission Internationale de l'Eclairage's (CIE) method for calculating colour coefficients was used to compute spectral-reflectance measurements over the visible wavelength range (CIE, 1931). According to the CIE colour system, coefficients were computed by a given colour being represented by x, y and z which contain hue and saturation data. These x, y and z coefficients identify colour differences between samples with y representing brightness, and x and z being virtual components of the primary spectra (Barthod et al., 2015). The x and y identify colour differences from blue to red and blue to green, L represents brightness and a, b, u, and v represent the chromaticity coordinates as opponent red-green and blue-yellow scales (Rossel et al., 2006; Barthod et al., 2015). MATLAB was used to convert the x, y and z coefficients into useable RGB colour values (Barthod et al., 2015; Westland et al., 2012). RGB colour coefficients were calculated from the spectral-reflectance by averaging reflectance data corresponding to the blue, green and red Landsat bands (Martinez-Carreras et al., 2010). Lastly, the averaged reflectance data were multiplied by 255 to obtain the eight bit pixel colour resulted in 16 calculated colour coefficients presented in Table 2.2 (Rossel et al., 2006; Barthod et al., 2015).

Table 2.2 Sixteen spectral-reflectance colour coefficients derived from a spectroradiometer using MATLAB and adapted from Martinez-Carreras et al. (2010)

Colour Space Model	Parameter	Parameter Abbreviation
RGB	Red	R
	Green	G
	Blue	B
CIE xyY	Chromatic coordinate x	x
	Chromatic coordinate y	y
	Brightness	Y
CIE XYZ	Virtual component X	X
	Virtual component Z	Z
Decorrelated RGB	Light intensity (brightness)	L
CIELAB	C.c. opponent red-green scales (chromaticity)	A
	C.c. opponent blue-yellow scales (chromaticity)	b
CIELUV	C.c.a. opponent red-green scales (chromaticity)	U
	C.c.a opponent blue-yellow scales (chromaticity)	V
CIELCH	CIE hue	C
	CIE chroma	H

2.2.4.3 Particle size and shape image analysis

Particle size and shape were measured using an image analysis technique. Image analysis instruments are typically equipped with multiple cameras that capture detailed pictures of the sediments as they are transported between the camera's lenses. The entire measuring range is very accurate with a great deal of sharpness due to these instruments typically consisting of a two-camera system. Computer software is used to identify size and shape characteristics by utilizing these images. Many studies including Roussillon et al. (2009), Miller and Henderson

(2010), and Liu (2014) have used image analysis to define the morphological features of particles.

The image analysis instrument used to analyze samples from BBW was the Sympatec, LIXELL as a dispersing unit for QICPIC software (Sympatec, Potato Research Center, Agriculture and Agri-Food Canada). The Sympatec LIXELL unit, however, was only capable of measuring particles ranging from 2 µm to 600 pm, therefore clay size fractions were not measured. Particles were introduced to the measuring zone through a the LIXELL (liquid cell) wet dispersing unit. Particles flowed past digital (CCD) cameras; multiple images were taken and then analyzed to assess the size and morphology of the subsample using QICPIC sensor software. Only a small sample (i.e., < 5 g) of soil was required for analysis and a dispersing additive was added to mitigate static interactions and increase the flow ability of the sample for improved image analysis. The subsample was transported to the measurement zone by a plastic tube feeder where the particles traveled between a black light and two cameras. Three separate outputs were produced as well as a fourth output representing the overall average. The primary results were based on number distributions including particle volume, area and length for size and sphericity, convexity and aspect for shape.

2.2.5 Statistical analysis

Several statistical tests were used, including: canonical discriminant function analysis, Shapiro-Wilk test, biplot analysis, Kruskal-Wallis test and stepwise discriminant function analysis to select the signatures capable of discriminating potential sources of suspended sediments in BBW. Multivariate fingerprinting relies on testing a large suite of potential signatures and statistical selection to determine the optimum suite of signatures that will best allocate sediments to individual sources (Davis and Fox, 2009). The process for statistical selection of fingerprints is depicted in the flowchart presented in Fig. 2.5.

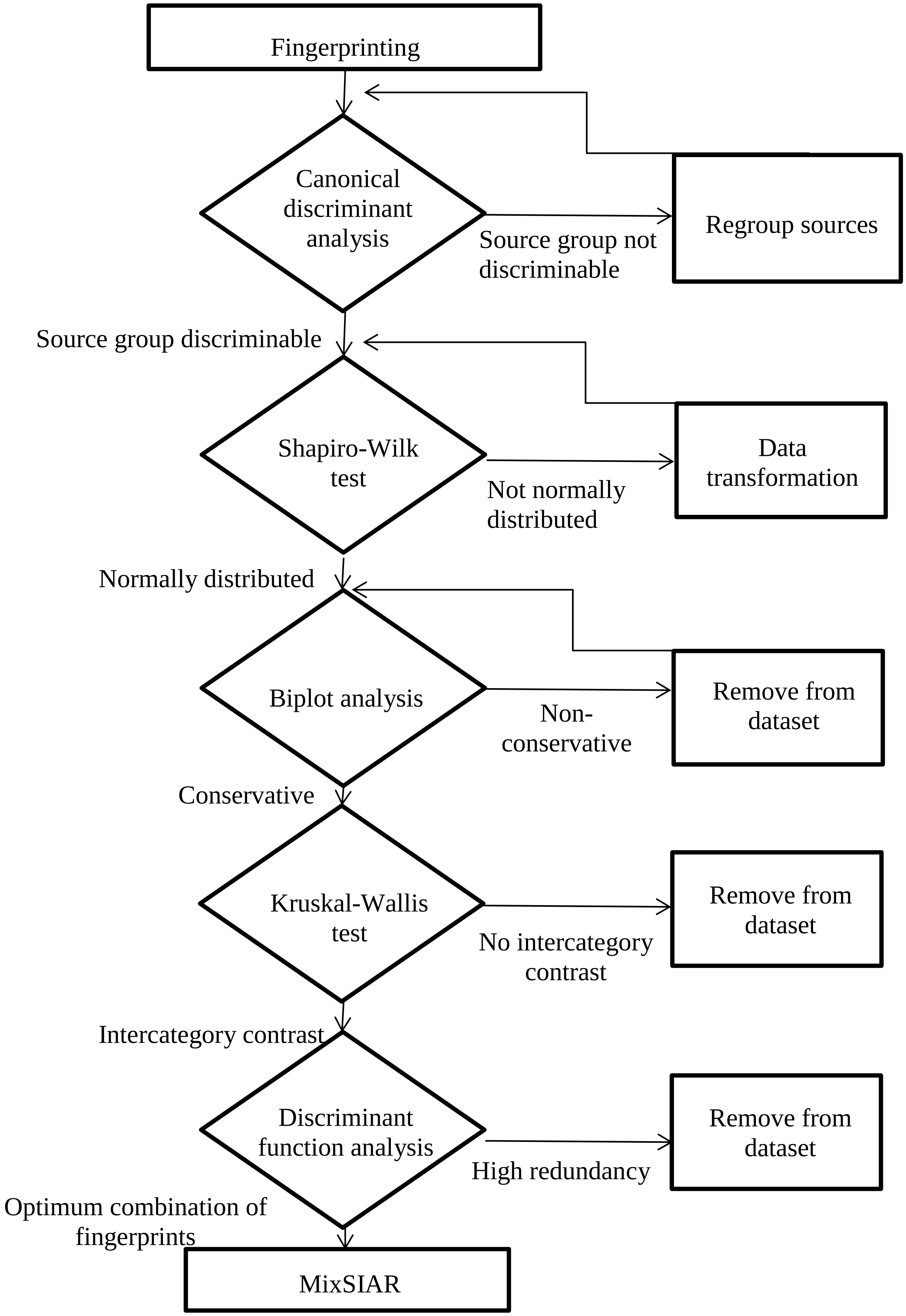

Figure 2.5 Flowchart for statistical selection of fingerprinting signatures utilized for sediment fingerprinting studies.

2.2.5.1 Canonical discriminant analysis

The canonical discriminant analysis is used in fingerprinting studies to determine the optimal separation of groups and remove fingerprints based on certain linear transformations. Reclassification and downsizing of original source groups is often necessary for efficient source type apportionment in fingerprinting studies (Walden et al., 1997; Barthod et al., 2015). Combining sources with similar fingerprints enables the minimum number of source groups to be utilized without valuable fingerprinting data to be lost. As the number of potential sources included in a mixing model increases, the uncertainty in the contribution of any one source also increases and, therefore, acquiring a minimum number of sources without missing any potential sources is required when using models such as MixSIAR (Nosrati et al., 2014). The canonical discriminant analysis emphasizes the dimensionality present in the dataset which allows for the proper classification of sources (Barthod et al., 2015). If the dataset lacks dimensionality, meaning that the creation of logical source groups is not possible, it may be necessary to replace fingerprints with alternate signatures that are better able to produce discriminable source groups.

Given a nominal variable (sediment sources) and several interval variables (sediment properties) canonical discriminant analysis transforms the original data to derive canonical scores. Each variable (sediment source) which is identified by a nominal classification label, is assigned two canonical scores based on its associated interval variable (sediment properties). The first canonical score represents the maximum multiple correlation as derived by a linear combination of variables that have the highest possible multiple correlation. The second canonical score represents the linear combination uncorrelated with the first canonical variable that has the highest possible multiple correlation with the groups. The resulting canonical scores are plotted on a scatter plot graph so the source groups are easily assessable.

2.2.5.2 Shapiro-Wilk test

Source and sediment data were tested for normality using the Shapiro-Wilk test. Fingerprinting data are required to be normally distributed because the mixing model used for apportioning sources, MixSIAR, assumes that all data are normally distributed. Violating the assumption of normality may result in inaccurate source apportionment estimates in MixSIAR. Therefore, all sediment and source data must be normally distributed in order to be utilized in apportionment modeling (Barthod et al., 2015). The Shapiro-Wilk test was performed in SAS and a 95 % significant level ($p < 0.05$) was used (SAS Institute Inc., 2013)

2.2.5.3 Biplot analysis

Biplot analysis provides a simple visual means of qualitatively evaluating the conservativeness of multiple variables and validating that all major sources have been accounted for (Smith and Blake, 2013). Many physical signatures are often identified as being non-conservative due to changes during transport and storage. There is, therefore, a need to account for these changes through the use of technology or by alternate means. Accounting for the non-conservative behaviour of sediments, however, is difficult due to the complex processes surrounding sediment dynamics. Therefore, biplot analysis represents an important step in the selection of fingerprints, as it identifies signatures showing non-conservative behaviour.

The biplot analysis was performed in MixSIAR through open source statistical software R package (R Package, 2012). The analysis produces Isospace plots that plot suspended sediment samples against the means and standard deviations for all the potential source material. Source means are represented by a center point and standard deviations are represented by error bars which extended from the source mean (±1 SD). Long error bars indicate substantial variability and uncertainty among data. Isospace plots were produced to determine whether the suspended sediment samples were within the range of the source material, suggesting that fingerprinting signatures were conservative and all major potential sources were included (Barthod et al., 2015). Suspended sediment samples plotted outside the standard deviations were considered to be beyond the potential source range resulting in the removal of the associated signatures due to non-conservative behaviour. Through the use of Isospace plots, the robustness of the composition of suspended sediments during storage and transport were assessed and used to select the most conservative signatures for BBW. Biplot analysis is considered the only statistical means of testing the conservativeness of fingerprinting signatures and, therefore, represents an important component of the model signature selection process (Smith and Blake, 2014).

2.2.5.4 Kruskal-Wallis test

The Kruskal-Wallis test was used by Collins et al. (1998) to determine the uniqueness of sediment sources based on the selected signatures. The Kruskal-Wallis test is a nonparametric procedure used to identify fingerprinting signatures that do not demonstrate a significant difference ($p > 0.05$) between source types (Walling et al., 1999; Collins and Walling, 2002). It is a test used to examine the existence of intercategory contrasts among source samples (Collins and Walling, 2002). The signature values for each source are compared to the same signature values for all other sources to determine if the differences in signature values are statistically significant. Kruskal-Wallis tests assess the null hypothesis which states signatures demonstrate no significant difference between source categories. Greater variation between categories thereby lowers the p-value. The Kruskal-Wallis test is the final step used in many sediment fingerprinting analyses in the classification of sources using the signatures destined for discriminant function analysis (Collins et al., 1998). In this study, the Kruskal-Wallis test ($p < 0.01$) was performed in open source statistical software R package (R package, 2012).

2.2.5.5 Stepwise discriminant function analysis

A stepwise discriminant function analysis was performed on normally distributed signatures which passed all prior statistical analyses. The stepwise discriminant function analysis was used to select a composite of fingerprints for MixSIAR by investigating the discrimination power of each signature and determining the number of samples correctly classified into each source group (Barthod et al., 2015). This analysis identified the optimal composite fingerprint that would result in the minimum number of signature properties necessary to provide the greatest discrimination between source materials without any repetition (Collins and Walling, 2002). The identification of an optimum fingerprint is based on the minimization of the Wilks' lambda value (Collins and Walling, 2002). As signatures are added and removed from the analysis, the variability within the source categories is reduced relative to the variability between categories, resulting in the lambda value to approach zero (Smith and Blake, 2014). In this study, the discriminant function analysis was performed in open source statistical software R package (R package, 2012). The end result of the stepwise discriminant function analysis was used in MixSIAR to estimate sediment source apportionment.

2.2.6 MixSIAR

Source contributions to instream suspended sediments were estimated for SW1 using a multivariate mixing model, MixSIAR, developed by the creators behind SIAR and MixSIR (Stock and Semmens, 2013). MixSIAR is an ecological mixing model, originally designed for food-web and predator-prey relationships; it is however, a basic mixing model which can be applied to other environmental studies such as sediment fingerprinting as long as the assumption of linear mixing is met (Stock and Semmens, 2013).

MixSIAR is a fully Bayesian isotopic mixing model which uses probability distributions in its interpretation of source contributions (Moore and Semmens, 2008; Nosrati et al., 2014). Three main steps make up the Bayesian framework: 1) resolution of prior probability distributions for the model parameters, 2) creation of a likelihood function, and 3) creation of the posterior probability distribution for parameters using Bayes rule and the observed data to alter the prior distribution (Bolstad, 2007; Nosrati et al., 2014). Bayesian frameworks are able to incorporate large source numbers, concentration dependent variables and uncertainties which previous models such as ISOSOURCE lacked, making it an effective modeling tool for sediment source apportionment due to many fingerprinting studies incorporating many potential sediment sources and requiring uncertainty estimates (Parnell et al., 2013). Covariates such as time and site are also available, which are thought to influence sediment source apportionment. In some cases both the sediments and the sediment sources are expected to be a function of the covariates.

The statistical sampling method used by MixSIAR to approximate sediment source contributions is Monte Carlo. The Monte Carlo method is able to propagate uncertainty in model inputs into uncertainties in model outputs (Smith and Blake, 2014). The Monte Carlo method estimates source proportions (i.e., means and variances) by producing multiple simulations with random sampling of input variables and model parameters (Nosrati et al., 2014). The model fitting framework, used by MixSIAR, is a class of the Monte Carlo that uses the Markov Chain Monte Carlo (MCMC) and the Gibbs Sampler algorithm. The MCMC simulates draws that are slightly dependent and are approximated from a posterior distribution and involve repeatedly guessing values of sediment source apportionment (Parnell et al., 2010; Phillips et al., 2014). The basic idea behind the Gibbs sampling algorithm is that rather than probabilistically picking the next contribution estimates all at once, separate probabilistic choices for each selection are made (Gibbs et al., 1971). As sediment source proportions are estimated those values that are not probabilistically consistent with the data are removed. A Markov chain is created due to the requirement that the new estimates be similar to the older estimates (Phillips et al., 2014). Once MixSIAR has completed its run, samples of posterior sediment source proportions are generated.

2.3 Results

2.3.1 Canonical discriminant function analysis

The function Proc Candisc produced canonical scores that were plotted on a two-dimensional scatterplot in order to determine if sources were distinguishable. Canonical results for particle size and shape signatures are presented in Fig. 2.6.

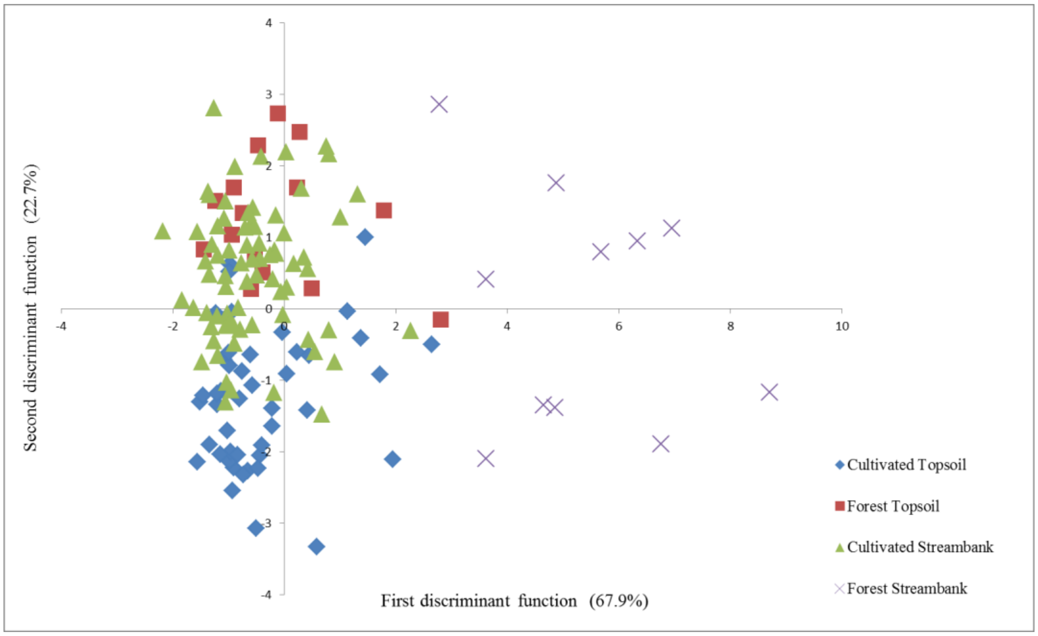

Figure 2.6 Canonical discriminant functions analysis scores for the discrimination of source groups using particle size and shape signatures for the following sediment source types: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambank, and 4) forest streambank.

The results of the canonical discriminant analysis conducted on the source samples show that the first two discriminant functions explain 91 % of the variability. The results which show the overlap in the plotted results for the sediment source types highlight the inability of particle size and shape to differentiate between the potential source types. This is likely due to a lack of dimensionality in the dataset (Barthod et al., 2015). In order to utilize particle size and shape signatures, reclassification and downsizing is required due to canonical scores for the following sources: 1) cultivated topsoil, 2) forest topsoil, and 3) cultivated streambanks plotting within the same general location on the scatterplot. Therefore, in order to use particle size and shape signatures as fingerprints, these three source types must be combined into one overall source type due to the inability of particle size and shape signatures to adequately discriminate these sources.

However, downsizing these source types (i.e., 1) cultivated topsoil, 2) forest topsoil and 3) cultivated streambanks) into one source type does not make sense for BBW and does not fit the overall objectives of the study and therefore it was decided to remove particle size and shape as potential signatures.

The results from the canonical discriminant analysis using Cs-137 and spectral-reflectance colour coefficients fingerprints are presented in Fig. 2.7. According to the canonical discriminant analysis, 81 % of the overall variability after source aggregation was explained by the first two discriminant functions. Approximately 55 % of the total variability was explained by the first discriminant function and 27 % was explained by the second discriminant function. This suggests that by plotting the first discriminant function against the second that the majority of the source sample information can be achieved.

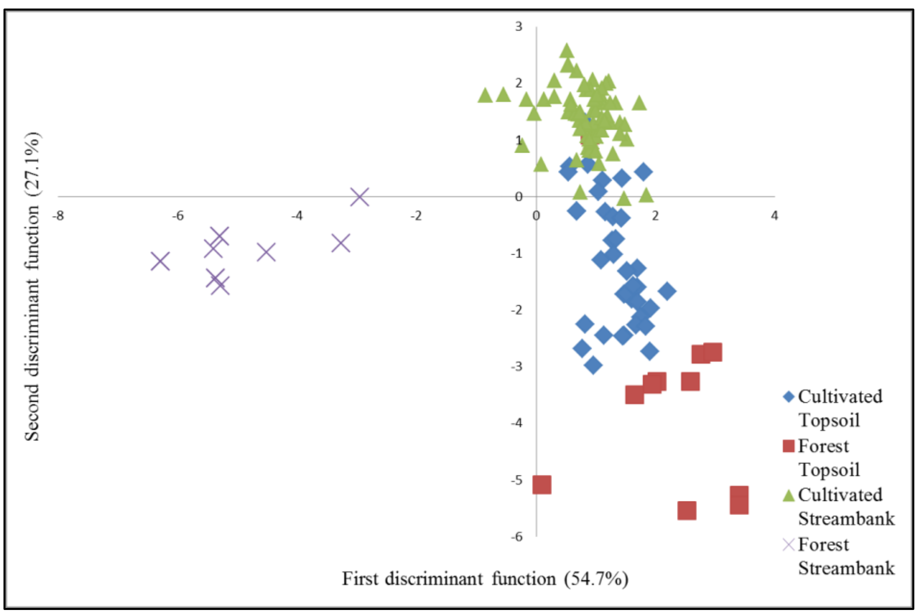

Figure 2.7 Canonical discriminant functions analysis scores for the discrimination of source groups using radionuclide Cs-137 and spectral-reflectance colour coefficients for the following sediment source types: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambank, and 4) forest streambank.

The plotted canonical scores depict that all sediment source types in BBW were relatively easily distinguishable. Results from the canonical discriminant analysis suggest that the majority of samples representing each source type lie within the range that allows each source type to be easily identifiable by the selected fingerprints. Therefore source types: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambank, and 4) forest streambank, were utilized as potential sources of suspended sediments in BBW.

2.3.2 Shapiro-Wilk test

Using the Shapiro-Wilk test, signatures were removed from further analysis if their associated source groups contained p-values greater than 0.05. Spectral-reflectance colour coefficient *A* and *C* (*C.c. opponent red-green scales and CIE hue*) did not pass the Shapiro-Wilk normality test and were therefore removed from further analysis.

2.3.3 MixSIAR biplot analysis

Eleven normally distributed colour coefficients (R, G, B, x, y, Y, X, Z, L, U, and H) and radionuclide Cs-137 were analyzed. Biplot results suggested removing colour coefficients x (chromatic coordinate x), y (chromatic coordinate y) and h (CIE chroma) from further analysis due to these colour coefficients producing erroneous results when paired with all other signatures.

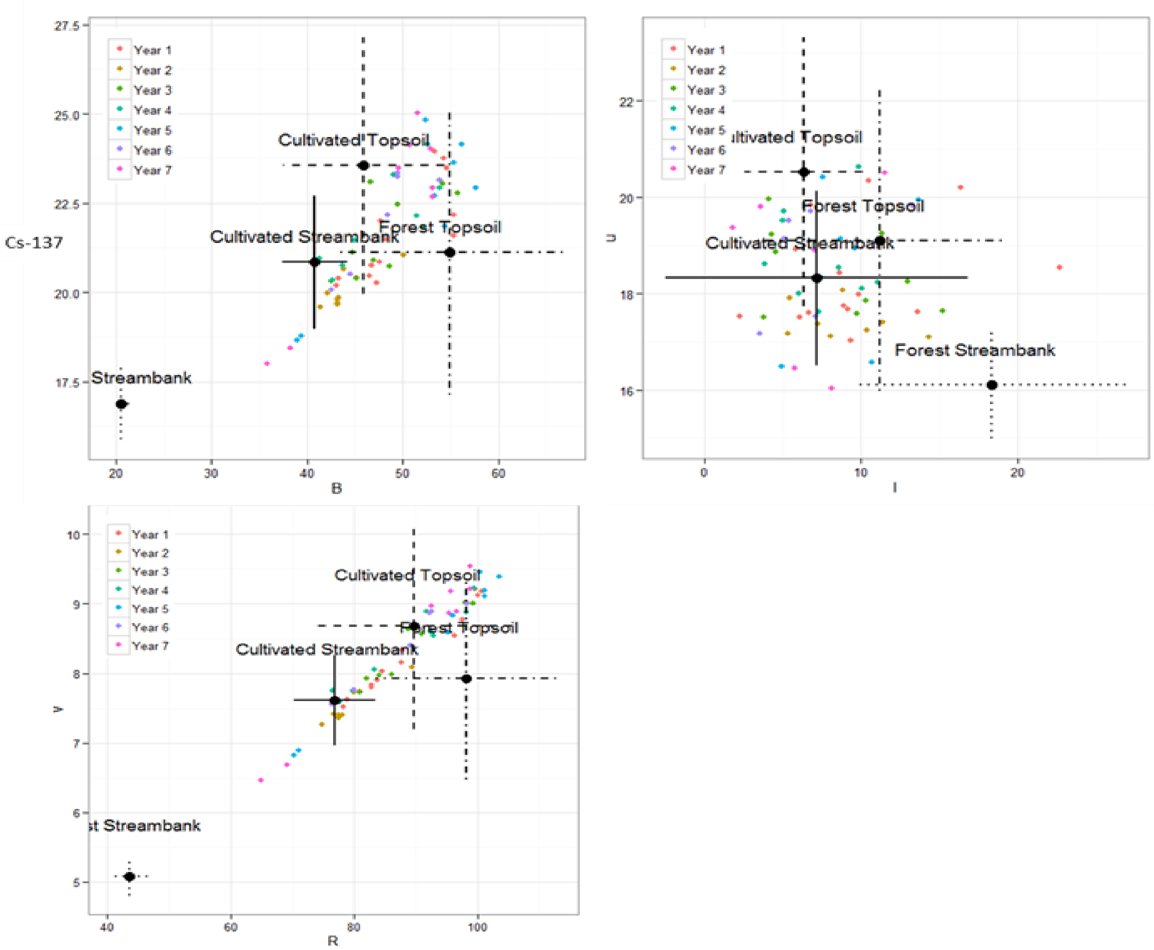

Figure 2.8. Isospace plots produced through biplot analysis showing the amount of uncertainty associated with the following sediment sources: 1) cultivated topsoil, 2) forested topsoil, 3) cultivated streambank, and 4) forested streambank.

Results from the MixSIAR biplot analysis suggest that cultivated topsoil and forest topsoil have the greatest amount of uncertainty, indicated by the long error bars in the Isospace plots as seen in Fig. 2.8. Isospace plots plot sources that are most similar, within close proximity; conversely those that are most different are plotted further apart. Results show that cultivated topsoil, forest topsoil and cultivated streambanks plot close together while forest streambanks are often plotted further away suggesting that forest streambank sources are more easily distinguishable compared to the other three sources (Fig. 2.8). In other words, the selected fingerprints are more capable of discriminating between forest streambank and the remaining three sources, as compared to between the other three sources.

2.3.4 Kruskal-Wallis test

The results from the Kruskal-Wallis test provided in Table 2.3 indicate that each fingerprinting signature was difference in fingerprint property between at least two of the four source types at a 99 % confidence interval ($p < 0.01$). Results suggest that spectral-reflectance colour coefficients and Cs-137 show inter-category contrast, and thus the hypothesis of stochastic homogeneity can be rejected and signatures can proceed to further analyses (Ruxton and Beauchamp, 2008; Barthod et al., 2015).

2.3.5 Optimum combination of fingerprints

Stepwise discriminant function analysis was employed as the final measure of source discrimination using fingerprints that passed all prior tests. Eleven variables (Cs-137, R, G, B, X, Y, Z, L, b, U, V) were included in the stepwise discriminant function analysis; nine signatures including V, R, L, U, B, G, Cs-137, X, and Z were indicated as being the optimum combination for discriminating sources in BBW. Table 2.3 and 2.4 shows the selected fingerprints and the percentage of source samples correctly identified by using the optimum combination of fingerprints.

Table 2.3 Results of the Kruskal-Wallis test and the stepwise discriminant function analysis applied to Cs-137 and spectral-reflectance colour coefficients: R, G, B, X, Y, Z, L, b, U, V

		Percentage of samples correctly classified				
Fingerprint Property	Kruskal-Wallis	Total	Cultivated Topsoil (%)	Forest Topsoil (%)	Cultivated Streambank (%)	Forest Streambank (%)
Cs-137	<0.01	35.6	72.7	37.5	33.8	0
R	<0.01	68.4	70.5	0	93	0
G	<0.01	69	70.5	0	94.4	0
B	<0.01	65	47.7	12.5	95.8	0
X	<0.01	67.8	68.2	0	93	0
Y	<0.01	69	70.5	0	94.4	0
Z	<0.01	65	45.5	18.8	95.8	0
L	<0.01	67.8	68.2	0	93	0
B	<0.01	65	59.1	0	83.1	50
U	<0.01	54	56.8	0	74.6	55.5
V	<0.01	66.7	61.3	0	87.3	50

Table 2.4 Results of the stepwise discriminant function analysis used to select the final combination of fingerprint properties essential for maximum source discrimination for potential source material: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambank, and 4) forest streambank

		Percentage of samples correctly classified				
Fingerprint Property	Fingerprint Combination	Total	Cultivated Topsoil (%)	Forest Topsoil (%)	Cultivated Streambank (%)	Forest Streambank (%)
V	V	66.7	61.3	0	87.3	50
R	V + R	72.4	63.6	37.5	90.1	50
L	V + R + L	70.1	66	37.5	83.1	50
U	V + R + L + U	75.3	68.2	43.8	88.7	77.8
B	V + R + L + U + B	76.4	68.2	43.8	90.1	77.8
G	V + R + L + U + B + G	79.9	68.2	50	90.1	94.4
Cs-137	V + R + L + U + B + G + Cs-137	81.9	72.7	62.5	88.7	94.4
X	V + R + L + U + B + G + Cs-137+ X	81.9	72.7	62.5	88.7	94.4
Z	V + R + L + U + B + G + Cs-137+ X + Z	81.9	72.7	62.5	88.7	94.4

The optimal combination of signatures produced an overall correctly classified percentage of 82 %; the associated correctly classified source type percentages were as follows: 73 % for cultivated topsoil, 63 % for forest topsoil, 89 % for cultivated streambanks and 94 % for forest streambanks. The stepwise discriminant function analysis results show that selected signatures represent forest streambank and cultivated streambank materials exceptionally well with a correctly classified percentage of 94 % and 89 % respectively. Conversely, cultivated topsoil and forest topsoil contained the lowest correctly classified percentages at 73 % and 63 % which supports the biplot analyses' findings of large discrimination uncertainty ±1 SD for these potential sources.

2.3.6 Sediment source apportionment percentages

The optimum combinations of fingerprints selected by the stepwise discriminant function analysis (V, R, L, U, B, G, Cs-137, X and Z) were used in MixSIAR. The source apportionment results from MixSIAR including the uncertainty assessment are included in Fig. 2.9

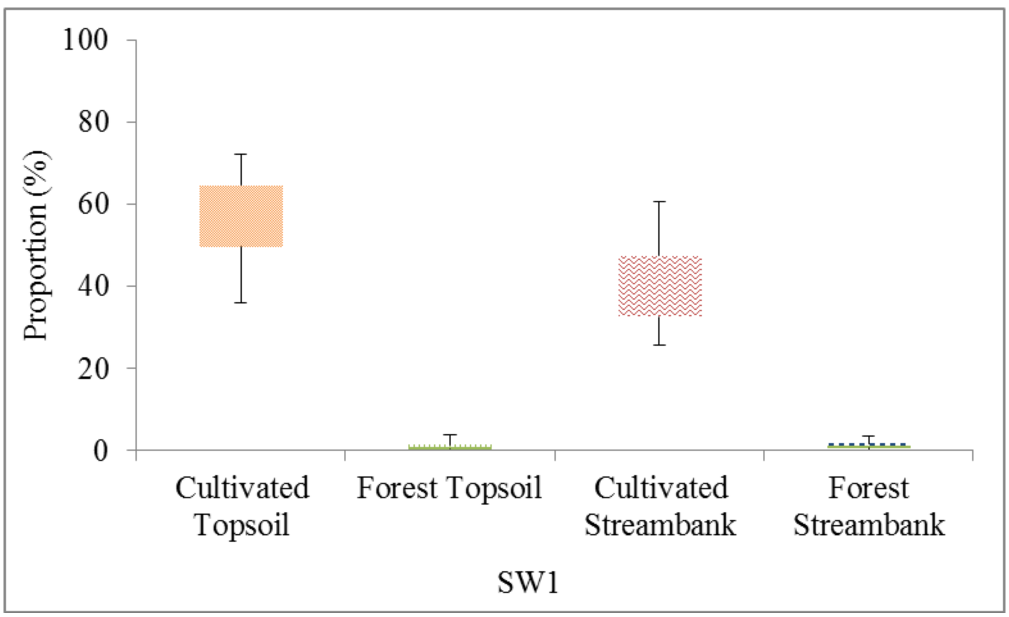

Figure 2.9 Box and whisker plot showing sediment source apportionment for SW1 using the following sources: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambank, and 4) forest streambank.

The results were reported as credibility intervals ranging between 25th and 75th percentiles. The box and whisker plots of the Monte Carlo results provide information on the uncertainty in the model results (Barthod et al., 2015). An appreciation for the variability can be determined by observing the distance between the first and third quartiles which suggest that cultivated topsoil and cultivated streambanks had the greatest variability which does not supports the biplot analysis findings that topsoil sources would have the highest variability. The low uncertainty assessments associated with forest topsoil and forest streambank sources are likely due to the low sediment source apportionment for these sources rather than these sources containing less uncertainty. At SW1, cultivated topsoil was found to be the predominant source of sediment, accounting for approximately 50 to 65 %. The model identified cultivated streambank as accounting for approximately 33 to 47 %. Forest streambanks were estimated to account for approximately 1 to 2 %. Forest topsoil was found to contribute the smallest amount of sediments, accounting for approximately 0 to 2 %. Literature from Saini and Grant (1980), Chow et al. (1999), and Chow et al. (2011) support these findings, reporting that cultivated land within this region is prone to intensive soil erosion. Therefore, the overall estimate of cultivated topsoil and cultivated streambanks being the main contributor to suspended sediments meets the general expectation for this catchment due to many studies identifying the intensive agricultural activities on soils susceptible to erosion as being the main cause of soil degradation and sediment loading within waterways in the region.

2.4 Discussion

2.4.1 Sediment source apportionment in BBW

A great amount of effort and funding has been spent on implementing BMPs in BBW, specifically flow diversion terraces which have been implemented on more than 50 % of cultivated fields (Yang et al., 2009). These BMPs are intended to combat against upland erosion created by farming practices. Although results from the current sediment fingerprinting study identified cultivated topsoil as the main contributor of suspended sediments in BBW, the study also found that cultivated streambanks were a substantial contributor to suspended sediments. Therefore many of the assumptions made from previous studies are accurate in their identification of upland erosion as a leading source of suspended sediments. Little acknowledgment has been made, however, to the contribution that cultivated streambanks have on the creation of suspended sediments, which according to the current study is substantial. Therefore the implementation of flow diversion terraces is undoubtedly effective at combating much of the upland erosion; however a combination of BMPs to target both upland erosion (e.g., flow diversion terraces) and streambank erosion (e.g., vegetated buffer zones) would likely be more effective than the implementation of flow diversion terraces alone. Similar to the results found in BBW, many other fingerprinting studies have concluded that streambank sources were a significant source of suspended sediments. For example, a study in the South Tobacco Creek watershed in southern Manitoba conducted by Koiter et al. (2013b) was able to discriminate between three sources, including: 1) shale bedrock, 2) topsoil, and 3) streambanks. Sources of sediments varied with respect to location, where sediment sources in the upper region were dominated by topsoil sources and streambank and shale bedrock sources were dominate in the lower portions. Gellis and Noe (2013) examined sediment sources in the Linganore Creek Watershed, Maryland, USA, which drains the highest sediment yield region of the Chesapeake Bay watershed. Streambanks and cultivated land were found to contribute the most fine-grained sediment. The study also found that sources from streambanks occurred during high flows and in winter months. Furthermore, a study conducted by Voli et al. (2013) in four watersheds draining a municipal water supply reservoir in the Neuse River, North Carolina, USA found that in three of the four watersheds investigated, streambank erosion was the largest contributor of sediment.

The current study as well as the additional literature reviews proves the importance of the identification of predominant sediment sources prior to the implementation of BMPs or other management tools. This is especially true in watersheds such as BBW which have implemented, are maintaining and plan to implement agricultural BMPs on over half of cultivated fields. In watershed such as BBW, it may be beneficial to implement BMPs that are capable of reducing both upland and streambank sources of suspended sediments.

2.4.2 MixSIAR

Although the most recent Bayesian mixing models, such as MixSIAR, include many new features that have improved the overall output of mixing models, there is still much uncertainty surrounding these tools. The current study attempted to minimize uncertainty by: 1) ensuring all sediment sources were identified, 2) ensuring there were adequate differences among sources, 3) keeping the amount of sediment sources low, 4) meeting the minimum number of signatures (n-1), and 5) ensuring signatures were not correlated to one another and were normally distributed. In BBW, many watershed visits and field observations were made to ensure no major potential sediment sources were overlooked, as this could cause bias in the estimate of source contributions, since source contributions must sum to 100 % (Phillips et al., 2014). Furthermore, the number of potential sediment sources identified in BBW was kept as low as possible without excluding major sources. This was based on research from Phillips et al. (2014) finding that once the number of sources reaches above 6 or 7, the discriminatory power of the model declines substantially. In order to keep source numbers low, without excluding any key sources, source types were combined if they were statistically and logically similar (Phillips et al., 2005).

2.4.3 Spatial framework weaknesses

Many fingerprinting studies have followed the same spatial framework as was implemented in BBW where suspended sediments are collected at the main outlet of a watershed and used to represent conclusions for the entire catchment (e.g., Poleto et al., 2009; Mukundan et al., 2010; Collins et al., 2012). Recently, the importance of carefully considering sediment sampling locations along river basins (e.g., Koiter et al., 2013b, Barthod et al., 2015) and between sub-watersheds (e.g., Brosinsky et al., 2014a) has been recognized. Sampling solely the watershed's main outlet for sediments may not fully capture temporal and spatial variations. It may inadequately capture variations in sediment availability and storage within watersheds. This is especially true for watersheds that are characterized by channel barriers, dis-connectivity issues and composed of very different sub-watersheds. For example a study conducted by Brosinsky et al. (2014b) found that watersheds composed of several sub-watersheds tend to create considerable variability in source apportionment. In relation to BBW, sediments collected at SW1 may have much different origins compared to suspended sediments collected elsewhere along Black Brook. Studies conducted in nested watersheds similar to BBW support this hypothesis as many studies have found (e.g., Koiter et al., 2013b and Barthod et al., 2015) that major sources of sediments switched from the headwaters to the outlet, while others (e.g., Mukundan et al., 2010 reviewed by Mckinley et al., 2013) have found slight differences in sediment source apportionment along the length of a stream. There is, therefore, a need for comparisons between sub-watersheds in order to assess important variability in the origin of fluvial suspended sediments in BBW.

2.5 Conclusion

In summary, the results of the sediment fingerprinting study in BBW based on seven years (2008-2014) of sediment sampling demonstrates high amounts of sediment contributions from cultivated topsoil and cultivated streambank sources compared to other potential sources. The results suggest that in BBW the main sources of sediment include cultivated topsoil (50 to 65 %) followed by cultivated streambanks (33 to 47 %), forest streambank (1 to 2 %) and forested topsoil (0 to 2 %) (25^{th}-75^{th} percentiles). This research utilized the capabilities of spectral-reflectance colour coefficients, in combination with a more conventional signature of Cs-137, to discriminate sediment sources at the main outlet of BBW. Although sediment fingerprinting represents a quick and relatively effective method to pinpointing sources of sediment, the spatial framework used in the current study (i.e., SFS utilizing Single Watershed Source Sampling) may create an incomplete assessment of sediment origins at a watershed scale. This may thereby create problems developing and targeting BMPs to minimize the impacts of agriculture on water quality if incorrect sources are identified as being the leading cause of suspended sediments. There is a need to address the shortcomings of the SFS to target non-point sources of suspended sediment.

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3. ASSESSMENT OF SEDIMENT SOURCE APPORTIONMENT UNDER DIFFERENT SPATIAL FRAMEWORKS FOR SEDIMENT SOURCE FINGERPRINTING STUDIES

Abstract

The sediment fingerprinting technique is increasingly being used to improve the understanding of sediment dynamics within watersheds. Many limitations are presently preventing the ability of this technique to trace sediments back to their sources. These limitations include the non-conservative nature of many sediment properties, connectivity, selectivity and spatial and temporal variation. Most of the literature on sediment fingerprinting tends to assume that there is a direct connection between sources and sinks, thereby resulting in the collection of suspended sediments at a single sampling location and conclusions based on these samples extended to represent sediment dynamics throughout the entire watershed. However, recent literature has described processes that occur between the sediment source locations and the point of collection downstream as a black box due to these processes not being well understood. In this study, we investigate sub-watersheds within a larger catchment we examined previously. The objective of Chapter 3 was to: 1) present additional spatial frameworks that may be used in complex (heterogeneous) watersheds that are characterized by varying geology, pedology, hydrology, vegetation cover and land management practices, requiring catchments to be broken down into different sections in order to better understand processes affecting sediment dynamics, and 2) determine which spatial framework best represents sediment dynamics at a watershed scale.

Spatial frameworks were divided into two groups: 1) Simple Fingerprinting Study and 2) Complex Fingerprinting Study. A Simple Fingerprinting Study (e.g., Chapter 2) treats each

watershed as a single catchment and assumes conditions affecting the production and transport, from land to stream, for each source type are uniform across the watershed. A Complex Fingerprinting Study assumes watersheds are heterogeneous; therefore catchments are broken down into different sections in order to better understand sediment processes. Various source sampling strategies were also created to complement the Simple and Complex Fingerprinting Study.

The results of Chapter 3 demonstrated the importance of spatial framework due to the source apportionment results differing with regards to the completeness at a watershed scale. Nested spatial frameworks demonstrated that there was a switch in sediment sources between the headwaters and the outlet of the watershed that was not detected in the non-nested spatial frameworks. This research highlights the importance of sampling location at a watershed scale especially with regards to the interpretation of sediment source apportionment results.

3.1 Introduction

Eroded sediments are important non-point source pollutants causing surface water degradation (Lamba, 2015). The loss of sediment-bound nutrients, such as phosphorus and nitrogen, from agricultural runoff into surface waters often results in the growth of toxic algal blooms and eutrophication. Eroded sediments are also a major source of contaminants such as pesticides in agricultural watersheds. In the Appalachian region of Atlantic Canada, many potato fields are prone to some of the most serious water erosion in Canada due to the climate, and topography conditions in the region and the intensive management associated with potato cropping. The eroded sediments end up in nearby water bodies causing water degradation.

With no clearly defined entry points, large land masses, high temporal and spatial variability and many stakeholders, it is challenging to pinpoint and manage this non-point source pollution (Koiter et al., 2013b). Sediment fingerprinting offers a potentially valuable tool to identify sediment sources. It utilizes a combination of field data, laboratory analyses and statistical modelling techniques in order to determine sediment source apportionment (Davis and Fox, 2009).

In the previous chapter, a Simple Fingerprinting Study (SFS) using a Single Watershed Source Sampling strategy was utilized to collect suspended sediments from a single target location (the main outlet) of the Black Brook Watershed (BBW) and all potential sediment source samples were collected from throughout the catchment. Suspended sediment samples were collected seasonally from 2008 to 2014 and sediment source samples were collected during two different campaigns in autumn 2009 and autumn 2014 and included the following potential sediment sources: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambank, and 4) forest streambank. Suspended sediment samples were collected at a single target location and results based on these samples were used to represent sediment dynamics throughout the entire watershed. Various fingerprinting signatures were measured in both suspended sediments and their potential sources. Cs-137 and spectral-reflectance colour coefficients were selected using statistical and processed-based selection criteria and ultimately used in the multivariate mixing model, MixSIAR, to estimate sediment source apportionment. The previous study concluded that the main sources of sediments accounting for approximately 50 to 65 % (25^{th}-75^{th} percentiles) of sediments were topsoil of cultivated land, while streambanks from cultivated areas, streambanks in forested areas and topsoil from forested areas accounted for approximately 33 to 47 %, 1 to 2 % and 0 to 2 % of sediments, respectively.

Multiple studies have suggested the importance of well-designed spatial frameworks (i.e., sediment and source sampling approaches) for the progression of the sediment fingerprinting technique (e.g., Koiter et al., 2013a; Mckinley et al., 2013). Within the sediment fingerprinting literature, the processes used to connect sources of sediments to the collected sediments have been described as a "black box" due to the lack of knowledge on processes occurring from the time inputs (i.e., sediment sources) are created to when the outputs (i.e., suspended sediments) are identified (Koiter et al., 2013b). As suggested by Koiter et al. (2013b), in order to overcome the problems associated with the black box, there needs to be an improvement in suspended sediment and sediment source sampling. Difficulties with spatial frameworks often arise due to sediment transport and storage complexities occurring at the watershed scale. These complexities make it difficult to understand the fate of eroded soils, and transport of the resulting sediments within the watershed. This is the result of many complex watershed variables such as soil, vegetation, climate, topography and human disturbances which often affect sediment dynamics (Davis and Fox, 2009). Studies including Walling (1983) and Koiter et al. (2013b) have noted that only a small proportion of eroded sediments within a river basin make it to the main outlet of a watershed. This has been attributed to sediment deposition within hillslopes, floodplains, river channels and/or other compartments of watersheds, which reduces connectivity (Koiter et al., 2013b). Therefore, with the likely possibility of dis-connectivity issues within watersheds, sampling suspended sediments solely at a watershed's main outlet may not completely represent processes affecting sediment dynamics elsewhere in the watershed. Furthermore, spatial and temporal variation can create significant variation in sediment apportionment throughout a watershed (Carter et al., 2003). Spatial and temporal variation in sediment properties is the premise that sediment fingerprinting is founded on, as it directly reflects variations in the relative contribution of sediment from distinguishable sources (Collins et al., 1997). However, the large variability of environmental variables over spatial and temporal scales is difficult to predict and model precisely making it difficult, especially in large catchments, to accurately estimate source contributions (Davis and Fox, 2009).

The objectives of this study were to: 1) extend our work in BBW by exploring additional spatial frameworks that may be utilized in complex, heterogeneous watersheds that require catchments to be broken down into different sections in order to better understand processes affecting sediment dynamics and compare the benefits and drawbacks of each spatial framework, and 2) Determine which spatial framework best represents sediment dynamics within the watershed. Spatial frameworks to be presented in this Chapter are intended to more fully capture variations in watershed characteristics including size, connectivity, transport and storage of sediments and changes in landuse, spatial and temporal variability and improve the interpretation and conclusions made from sediment fingerprinting studies.

3.2 Materials and Methods

3.2.1 Spatial Frameworks

Spatial frameworks must be designed to ensure that all potential, significant sources of sediment are adequately sampled. As discussed above, because dis-connectivity, spatial and temporal variations within watersheds have large influences on sediment dynamics, additional spatial framework strategies have been designed to adequately estimate sediment source apportionment. Spatial framework strategies can be divided into two main categories: 1) Simple Fingerprinting Study (SFS), and 2) Complex Fingerprinting Study (CFS). The SFS and CFS may be broken down into different sediment source sampling approaches which are intended to be selected by researchers based on watershed characteristics and research objectives.

There can be substantial spatial variation in soil properties adding an addition layer of complexity in predicting the properties of eroded sediments entering waterways (Koiter et al., 2013a). The total number of samples to approximate a source sample distribution remains an unsettled issue and criteria developed based on source areas for the source and soil type is a major need facing the fingerprinting technique today (David and Fox, 2009). According to Davis and Fox (2009) plowed agricultural soils will require fewer samples to obtain a sample distribution than a heterogeneous forest soil. There is, therefore, a requirement to determine a minimum number of samples needed to adequately characterize the mean and variance of soil properties. This minimum number of samples has not been described in any of the past sediment fingerprinting studies, to the best of our knowledge. The number of samples used in previous sediment fingerprinting studies has varied substantially, with some studies collecting as little as 165 individual soil samples (i.e., Mukundan et al., 2010) while other studies have collected as many as 349 soil samples (i.e., Smith and Blake, 2014). Due to there being no past literature recommending a minimum number of samples required to adequately describe the mean and variance of soil properties required for fingerprinting, Koiter et al. (2013a) and Davis and Fox (2009) identified that there is a need for standardized guidelines for sampling to improve soil representation. Past research from Koiter et al. (2013a) referred to a soil fertility study conducted by Kariuki et al. (2009) in which advice on the number of samples required to obtain a statistically representative measure of soil properties was given (i.e., 22 soil subsamples). The current study has referred to a fertility study conducted by Slevinsky et al. (2001 and 2002), which concluded that a minimum of 30 samples per source type is necessary in order to comment on the mean and the variance of most soil properties.

3.2.2 Simple Fingerprinting Study (SFS)

In SFS, a watershed is treated as a single catchment with all potential source samples collected to represent a single catchment of interest. Simple Fingerprinting Studies are not spatially specific as they do not consider the direct physical link between sediments and sediment sources. These studies assume that conditions affecting production and transport, from the land to the stream, for each source type are uniform across the watershed with regards to geology, pedology, hydrology, weather, vegetation cover and land management. SFS typically include specific source sampling strategies, which in the current review have been named Watershed Source Sampling and Regional Source Sampling. Watershed Source Sampling can be applied to watersheds with a single or multiple locations of interest for sediment source apportionment. Specifics on Watershed Source Sampling for single and/or multiple target locations and Regional Source Sampling are presented in subsequent sections.

3.2.2.1 Single Watershed Source Sampling

To conduct Single Watershed Source Sampling, potential source materials are collected in sampling areas located within the watershed boundaries, typically upstream of the location intended for sediment source apportionment (Fig. 3.1).

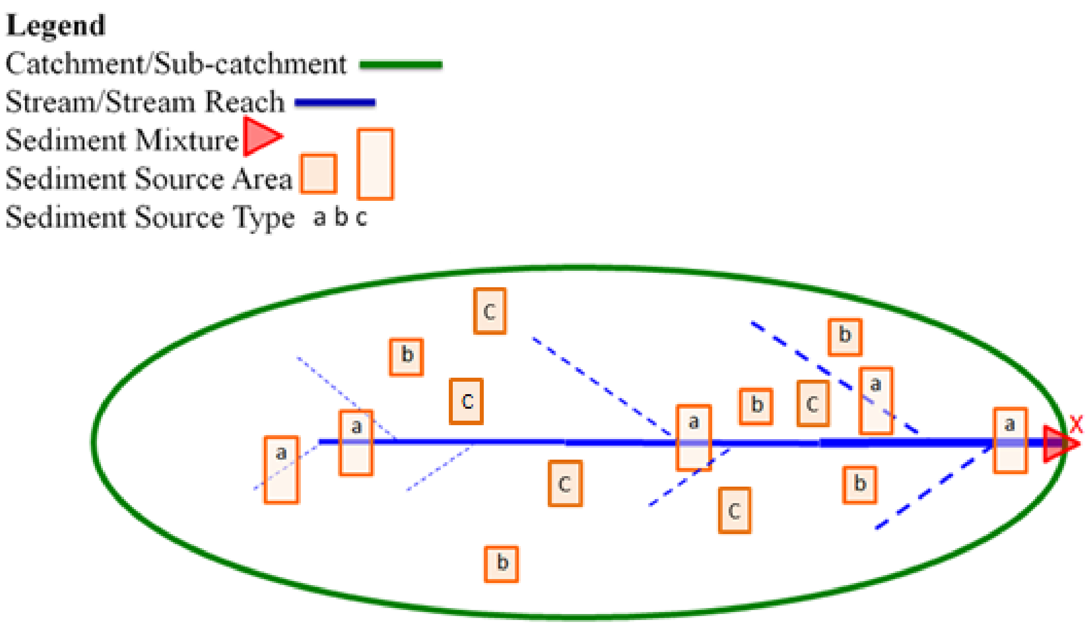

Figure 3.1 Single Watershed Source Sampling for a Simple Fingerprinting Study containing a single location of interest (X) for sediment source apportionment and three source types (a, b, c), each identified within five separate sediment source areas within the watershed of interest.

An example of how Single Watershed Source Sampling is utilized for a SFS containing one targeted area for sediment apportionment is illustrated in Fig. 3.1. Suspended sediments are collected at a watershed's main outlet (X) and all potential source materials are collected throughout the catchment in accessible areas, with visible signs of erosion and high connectivity to the channel of interest. Three potential source types (a, b, c) have been identified as contributing to suspended sediments collected at the main outlet (X). A total of five sediment source areas have been selected for each source type. A total of six source samples are therefore required to be collected from each source area to reach the recommended 30 samples per source type.

3.2.2.2 Regional Source Sampling

To conduct Regional Source Sampling, source materials are collected from across the watershed, however where there are not enough accessible areas to achieve an acceptable number of source samples within the catchment of interest, areas may be selected from outside the catchment, if there is reason to believe that they are representative of source material within the catchment undergoing sediment fingerprinting (Fig. 3.2).

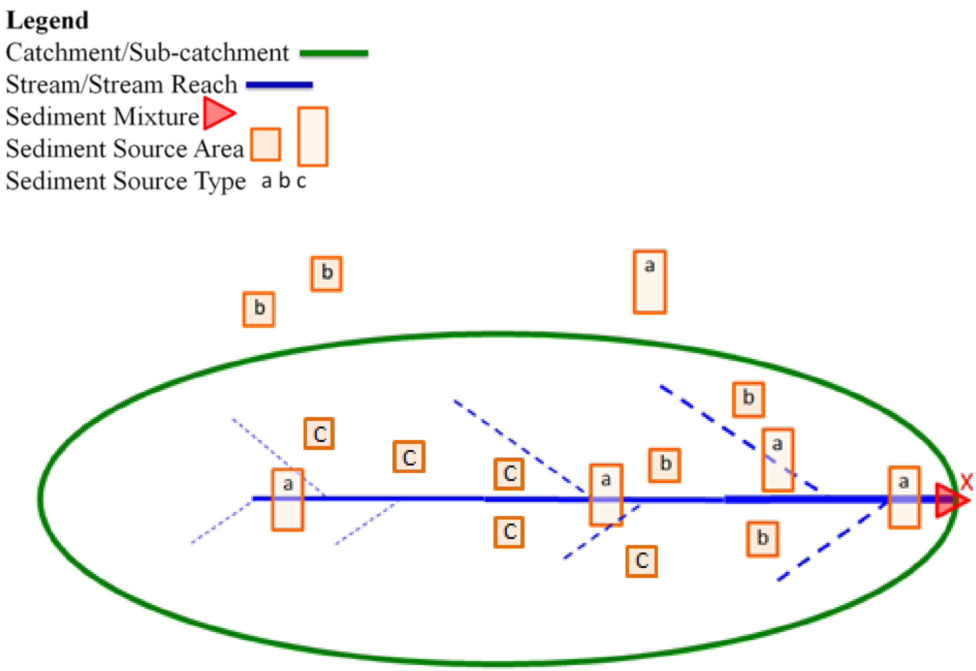

Figure 3.2 Regional Source Sampling for a Simple Fingerprinting Study where suspended sediments are collected at location X, with a, b and c identified as potential sediment sources collected in five sediment source areas from within and outside the watershed of interest.

An example of Regional Source Sampling utilized to determine sediment source apportionment for location X is depicted in Fig. 3.2. Potential sources (a, b, c) have been identified in five separate sediment source areas of which source type a and b have source areas located outside the watershed of interests. In this case, sampling outside watershed boundaries may be necessary to ensure source types contain an adequate number of samples to allow for proper characterization of the selected fingerprints. Regional Source Sampling may be considered when sampling conditions within the study watershed make it difficult to collect an adequate number of samples. Therefore in some situations acquiring an adequate amount of samples may require sampling outside the watershed of interest. Because SFS assume that conditions affecting production and transport, from the land to stream, for each source type are uniform within watershed boundaries, source samples collected outside watershed boundaries must be proven to be similar with regards to geology, pedology, hydrology, weather, vegetation cover and land management as samples collected within the watershed of interest.

3.2.2.3 Multiple Watershed Source Sampling

Simple Fingerprinting Studies may also contain multiple locations intended for sediment apportionment. Similar to Single Watershed Source Sampling, Multiple Watershed Source Sampling is characterized by source material collected from sampling areas within the watershed intended for apportionment. However, unlike a SFS containing one location, studies containing multiple sediment sampling locations may include sediment source material collected from sampling areas located both upstream or downstream of sediment sampling sites as potential sources. Using source samples collected from sampling areas located both upstream and downstream relative to sediment target locations may be advantageous because it increases sample numbers which is vital in accurately assessing the variance of soil properties. However, utilizing sediment source materials collected downstream of sediment collection locations requires the assumption that the conditions affecting production and transport for each source type are uniform across the watershed.

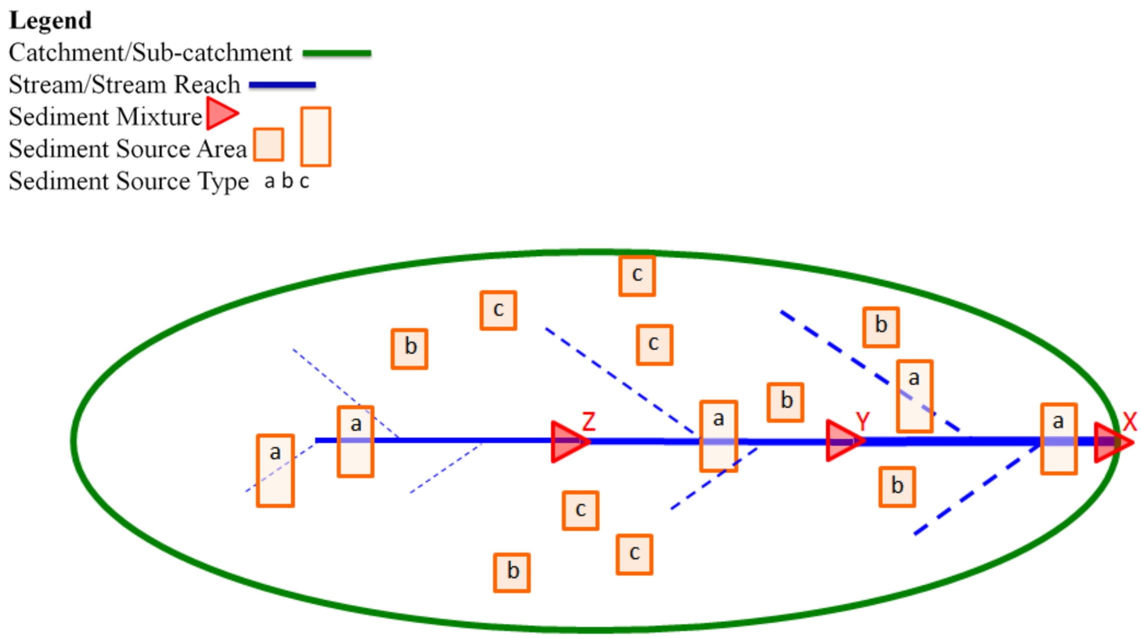

Figure 3.3 Watershed Source Sampling for a Simple Fingerprinting Study containing multiple sediment target locations (X, Y, Z) with three source types (a, b, c) identified and collected within five source areas within the watershed of interest.

An example of how Watershed Source Sampling is utilized for a SFS containing multiple sediment collection locations is illustrated in Fig 3.3. Suspended sediments are collected at multiple locations along a channel (X, Y, Z) and three potential source types (a, b, c) have been identified as contributing to suspended sediments at locations (X, Y, Z). Five sediment source areas have been selected for each of the three source types, requiring six source samples to be collected from each source area to reach the recommended 30 samples per source type. Because Watershed Source Sampling assumes that the catchment is homogeneous, all source samples, regardless of their location of collection, (upstream vs. downstream relative to target location), may be used to represent potential sediment sources for locations X, Y and Z.

3.2.3 Complex Fingerprinting Study (CFS)

Complex Fingerprinting Studies are typically used for more heterogeneous watersheds that require catchments to be broken down into different sections in order to better understand processes affecting sediment dynamics. These studies are spatially specific as they consider the direct physical link between sediments and sediment sources. These studies are utilized in watersheds characterized by varying geology, pedology, hydrology, vegetation cover and land management making it difficult to determine the origin of sediments collected in streams. Two source sampling strategies have been created to assist in determining sediment apportionment in CFS including: 1) Upstream Source Sampling, and 2) Reach Source Sampling.

3.2.3.1 Upstream Source Sampling

Upstream Source Sampling treats the watershed as multiple, nested catchments defined by sediment collection sites established at the outlet of several sub-watersheds. Sediment source materials are collected from sampling areas located from across the catchment, however only source material collected from sampling area located upstream of sediment collection locations are considered as potential sources of sediments. An example of the Upstream Source Sampling approach utilized for a CFS is depicted in Fig. 3.4.

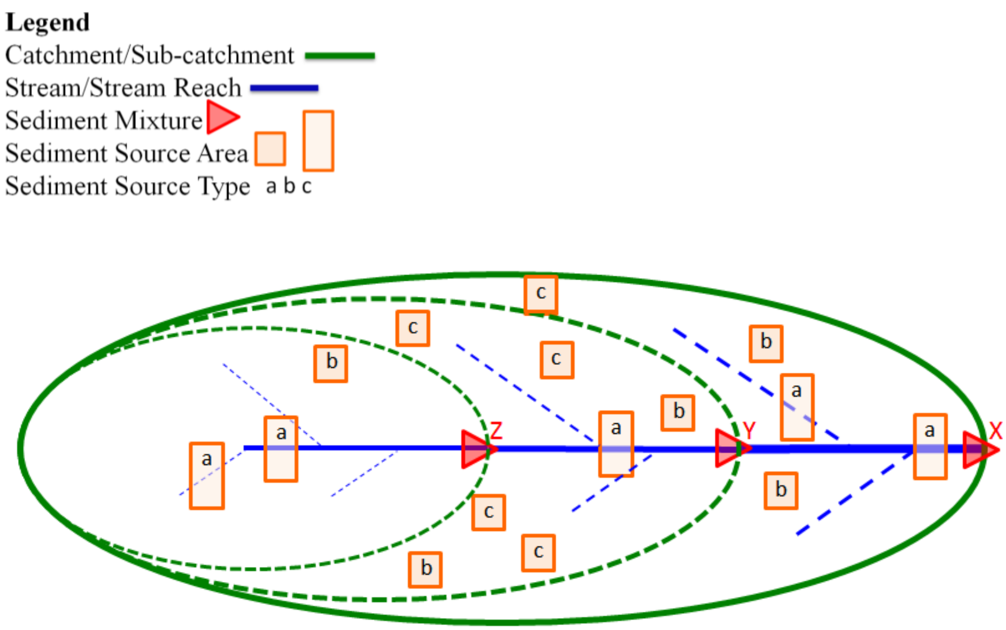

Figure 3.4 Upstream Source Sampling for a Complex Fingerprinting Study containing three nested sediment collection locations (X, Y, Z) and three potential source types (a, b, c) collected within five sediment source areas within the watershed of interest.

The sampling is exactly the same as the example shown for the SFS with multiple sampling locations. However, because Upstream Source Sampling only considers samples collected upstream as potential sediment sources, numbers of potential sources for sediment target locations (X, Y and Z) will vary. Location X (main outlet) will have the greatest amount of sediment source samples with: a=30, b=30 and c=30; location Y will have less sediment source samples with: a=18, b=24 and c=24 and location Z will have the least amount of sediment source samples with: a=12, b=6 and c=0. As sediment target locations move further upstream, it becomes more difficult to acquire the recommended amount of source samples (30) to adequately characterize the mean and variance of each signature property used in the fingerprinting analysis.

3.2.3.2 Reach Source Sampling

To conduct Reach Source Sampling, sediment source material must first be divided into local and non-local sources. Local sources are collected from areas with clear signs of erosion (i.e., topsoil and streambanks) typically within the sub-watershed in which suspended sediments are being collected for apportionment. Non-local sources are represented by suspended sediments collected within an upstream location in the stream of interest. Typically these sediments (i.e., non-local sources) are collected in a separate sub-watershed located upstream of the location for apportionment. These upstream suspended sediment sources are used to represent all potential non-local erosional soil sources contributing to suspended sediments at a target location in order to determine source proportion estimates. Reach Source Sampling treats the watershed as multiple nested sub-watersheds in order to reflect the difference between local and non-local sources contributing to suspended sediments.

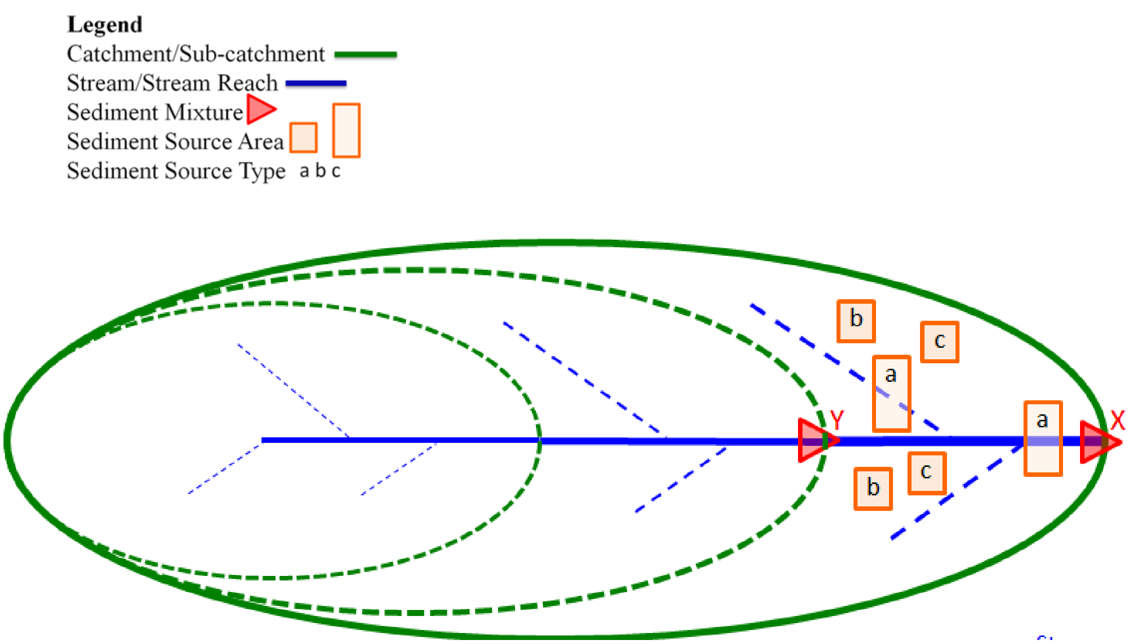

Figure 3.5 Reach Source Sampling for a Complex Fingerprinting Study containing one sediment collection location for sediment source apportionment (X) and three potential source types (a, b, c) as well as an upstream suspended sediment source (Y)

An example of Reach Source Sampling utilized within a CFS is illustrated in Fig. 3.5. Intensive local source sampling would be required within the nested sub-watershed of interest between the sediment apportionment location (X) and the upstream suspended sediment collection location (Y) which represents all non-local sources potentially contributing to suspended sediments. Three source types (a, b, c) have been identified within two sediment source areas, therefore a total of 15 samples are required to be collected from each source area to reach the recommended 30 samples per source type. Sediment source samples a, b and c are meant to represent local sources contributing to suspended sediments at location X. The value of examining the sediment at multiple locations allows for better understanding of how stream hydrology affects sediment transport and source identification at different scales.

3.2.4 Study Area

The present study was carried out in BBW which drains a 1,450 ha watershed (mostly cultivated) in northwestern New Brunswick (47°05' to 47°09'N and 67°43' to 67°48' W), located in the Appalachian region of Canada. BBW and the greater Little River Watershed (380 km^2) discharge into the Saint John River. These watersheds are part of the Upper Saint John River Valley Ecoregion in the Atlantic Maritime Ecozone (Marshall et al., 1999). The average annual temperature is 3.2°C (Environment Canada, 2012). The region has a mean annual precipitation of 1,134 mm, with approximately one-quarter of precipitation in this region falling as snow (Su et al., 2011). The climate is moderately cool boreal with a soil moisture regime ranging from humid to perhumid, with frost periods lasting for approximately 120 days (Mellerowicz et al., 1993). Topography in BBW consists of valleys along Black Brook with neighbouring plateaus. The elevation ranges from 180 to 260 m above sea level (Mellerowicz et al., 1993). The geological material of the area is mostly Ordovician and Silurian calcareous and argillaceous sedimentary rocks (shale, slate, limestone). Volcanic rocks also exist. The major glacial influence on the area resulted from the Wisconsin ice sheet. Surface deposits are glaciofluvial and morainal containing mixed sand, gravel, silt, and stones (; Langmaid et al., 1976, Langmaid et al., 1980, Mellerowicz et al., 1993).

BBW is comprised of eight nested sub-watersheds defined by water gauging stations (Fig. 3.6). Suspended sediments were collected at the outlets of four sub-watersheds (sub-watersheds 1, 2, 3, and 4) located along the main stem of Black Brook, along a longitudinal path, as well as an additional sub-watershed (sub-watershed 8) located on one of two tributaries that encompass Black Brook (sub-watershed 8). For simplicity, identification of sub-watersheds will follow the identification method in Chapter 2 with sub-watersheds 1, 2, 3, 4, and 8 represented by the following: SW1 (sub-watershed 1), SW2 (sub-watershed 2), SW3 (sub-watershed 3), SW4 (sub-watershed 4) and SW8 (sub-watershed 8). A nested sampling approach similar to the one used by Koiter et al. (2013b) was used to assess the influences of scale, connectivity and catchment characteristics including: hydrology, topography and geomorphology on sediment dynamics.

Soil erosion in this area of New Brunswick was not a factor prior to settlement due to complete forest cover, ranging from softwoods in the valleys, to mixed woods and pure tolerant hardwoods on the slopes and ridges. After settlement, soils in this region were recognized for their excellent qualities for mixed farming, and especially for potato cropping. Today, potato cropping is extensive, while the forests are absent in the most intensively managed areas with marginal areas containing some fragmented forests. The surrounding areas of BBW, however, are predominantly forested with some of these areas regularly being logged by local logging companies. The current landuse in BBW consists of predominantly agriculture throughout the watershed except for in the upper portion which has remained predominantly forested.

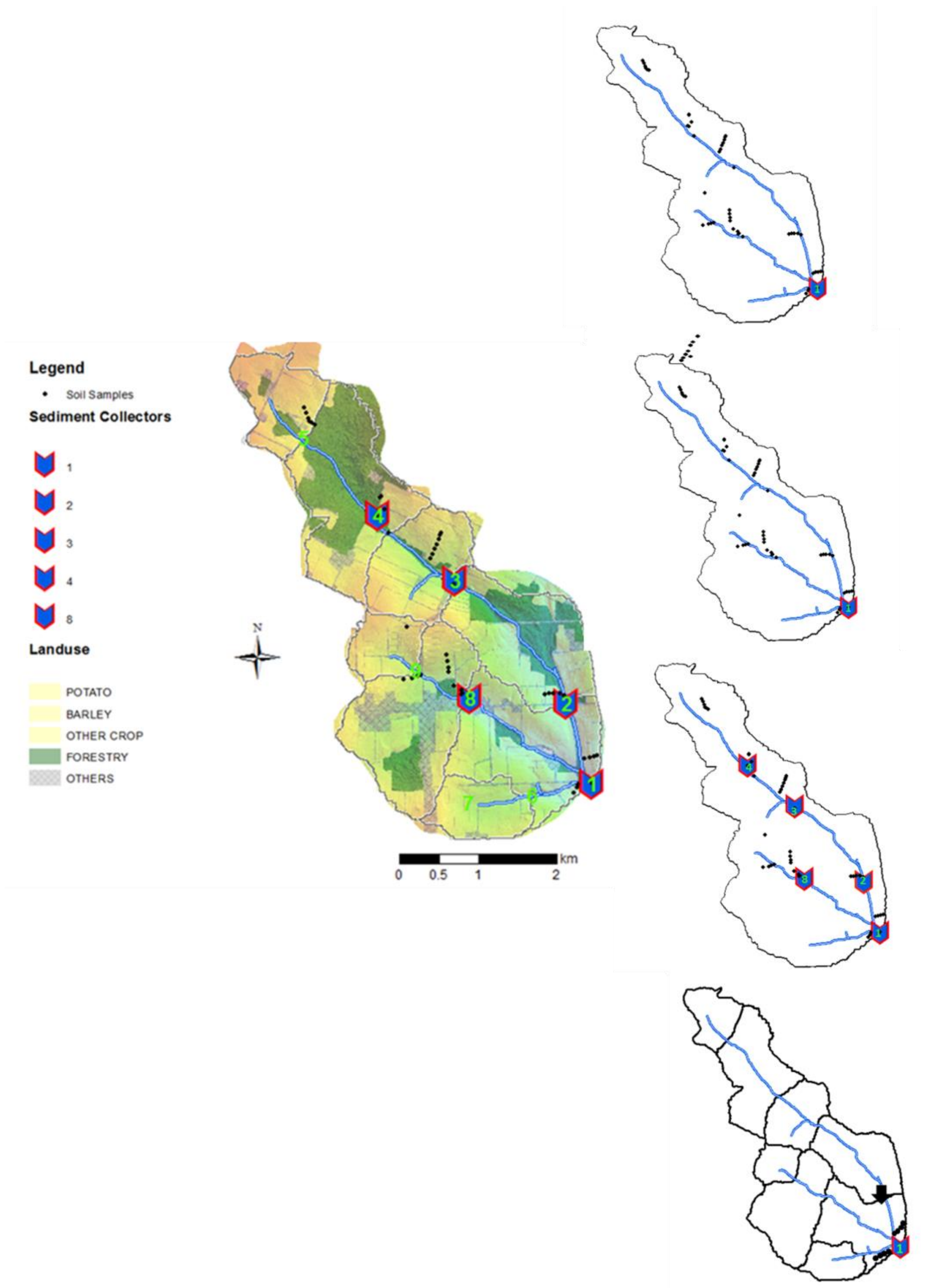

Figure 3.6 Black Brook Watershed located in the Appalachian region of Atlantic Canada, shown are the sub-watersheds, suspended sediment collection locations (SW1, SW2, SW3, SW4 and SW8), soil source sample locations and the watershed landuse.

Table 3.1 Local catchment areas and sub-watershed drainage areas and landuse percentages for sub-watersheds sampled for suspended sediments in BBW

Outlet station No.	Watershed drainage areas				Local catchment areas			
	Area (km^2)	Agriculture (%)	Forest (%)	Other (%)	Area (%)	Agriculture (%)	Forest (%)	Other (%)
SW1	14.4	65	21	14	2.4	77	7	16
SW2	8	44	43	13	2.3	55	36	9
SW3	5.6	40	46	7	2.4	77	12	11
SW4	3.2	14	65	21	3.2	14	65	21
SW8	3.3	66	13	21	3.3	66	13	21

In Table 3.1 landuse for the selected sub-watersheds has been divided into 3 categories: agriculture, forest and other. The agricultural land use category includes predominantly: potato, canola, corn, clover pasture and range land. The forested land use category includes a number of dominant tree species consisting of tolerant forests of: beech (*Fagus grandifolia*) and sugar maple *(Acer saccharum*) with scattered yellow birch (*Betula alleghaniensis*) found on the higher land and the slopes and valley bottoms dominated by balsam fir (*Abies balsamea),* white spruce (*Picea glauca)*, black spruce (*Picea mariana)* and white pine (*Pinus strobus*) (Loucks, 1962). The other landuse category includes residential areas, roads and wetlands.

3.2.5 Sampling

To assess the impacts of agricultural activities on soil erosion and the delivery of sediment to surface waters, source samples were categorized into four main potential source types. These potential sediment sources were: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambank, and 4) forest streambank. These four source types were utilized as potential sediment sources for: 1) SFS utilizing 1a) Watershed Source Sampling and 2 b) Regional Source Sampling, and for 2) CFS utilizing 2a) Upstream Source Sampling. However, potential sources of sediments differed for a CFS utilizing Reach Source Sampling with potential source materials including local sources made up of cultivated topsoil or cultivated streambank. Forest sources were not included as a potential local source of sediment due to Reach Source Sampling only being only applied to SW1, SW2 and SW3 which contain no local forest sources (all forest samples were collected upstream of SW4). Non-local sources were made-up of sediments collected upstream of the target location for apportionment and were intended to represent all potential erosional sources located in areas considered non-local (i.e., located outside the sub-watershed containing target area for apportionment).

The sampling technique utilized in the current study followed the same procedure employed in Chapter 2. In brief, potential sediment source material from topsoil and streambank sources were collected during two campaigns, with potential topsoil sources collected in October, 2009 and potential streambank sources collected in October, 2014. Both topsoil and streambank samples were collected based on knowledge of erosion susceptibility, accessibility and connectivity to the channel of interest. Spatially distributed soil samples from across BBW consisted of approximately 57 topsoil and 76 streambank samples collected from throughout the watershed. Source samples were also collected outside BBW in Little River watershed to provide additional forest source samples for Regional Source Sampling. In total, 3 forest topsoil and 10 forest streambank source samples were collected in Little River Watershed. The spatial location of sampling sites is shown in Fig. 3.6. As stated in section 2.2.3, topsoil samples were collected from the upper 0-5 cm depth of the soil using the transect sampling method and corers were used to obtain samples. Individual streambank samples were collected to form a full streambank profile by sampling beginning at the top of the streambank working downwards in 10 cm increments using a box-core sampler.

Suspended sediment samples were collected using Phillips et al. (2000) designed in-situ, time-integrated samplers (Fig 2.4). In-situ time-integrated samplers were installed at five locations along Black Brook including: BBW's main outlet (i.e., SW1) and the outlets of SW2, SW3, SW4 and SW8 (Fig. 3.6). Suspended sediments were collected seasonally, excluding winter, from 2008 to 2014. In order to collect the necessary amount of suspended sediments, two versions of the Phillips et al. (2000) designed in-situ time-integrated suspended sediment sampler were utilized. One version contained similar dimensions to the Phillips et al. (2000) design, while the other was a smaller version.

Table 3.2 Dimensions for the Phillips et al. (2000) in-situ time-integrated suspended sediment sampler and the dimensions for the smaller sized sampler used in BBW

Time-integrated sediment sampler	Dimensions for Phillips et al. (2000) sampler (cm)	Dimensions for BBW sampler (cm)
Internal cross section	9.8	7.4
Cylindrical tube length	100	62
Inlet tube internal cross section	0.4	0.4

The smaller samplers were intended for sections of the stream that experience low flows (i.e., SW2, SW3, SW4 and SW8) while the larger samplers, with nearly the same dimensions as the Phillips et al. (2000) design, were utilized in sections of the stream with high flows throughout the year (SW1). Table 3.2 contains the measurements of the Phillips et al. (2000) sampler for comparisons to the modified versions used in BBW (Table 3.2).

3.2.6 Signature selection and laboratory analysis

Because our preceding study had shown the ability of radionuclides and spectral-reflectance (colour) properties to discriminate sources, we decided to expand their use in this study. The analysis used an identical suite of initial fingerprinting signatures as utilized in

Chapter 2 including Cs-137 and 16 colour coefficients. Whereas, Cs-137-is an anthropogenic radioisotope that relies on atmospheric deposition, soil colour signatures rely on differences in organic matter, drainage, parent material and/or chemical/ biological weathering to differentiate sediment source origins. The pairing of fingerprinting signatures derived from different processes improves confidence in sediment source apportionment results with specific pairing of colour signatures with more conventional fingerprinting properties, such as Cs-137, recommended by Martinez-Carreras et al. (2010).

A high purity geranium gamma spectrometer (Canberra, Potato Research Centre, Agriculture and Agri-Food Canada) for the detection of Cs-137 (661.7 keV) and a 350-2500 nm wavelength range spectroradiometer (ASD FieldSpec Pro, Analytical Spectral Device Inc.) to compute spectra reflectance measurements (see. Chapter 2.2.4) were used to analyze selected fingerprint signatures.

3.2.7 Statistical analysis and multivariate modeling

The first step in the sediment fingerprinting process was the canonical discriminant analysis which was used to reclassify and downsize sediment sources into groups distinguishable from one another (Barthod et al., 2015). A normality test was performed on all source material using the Shapiro-Wilk test ($P < 0.05$) (SAS Institute Inc., 2013). The normality test was conducted to ensure the mixing model's assumption of normally distributed data was met (Stock and Semmens, 2013). Next, a biplot analysis (i.e., range test) was conducted by plotting the distribution of suspended sediments against the means and standard deviations of potential sediment sources. This was done to ensure that all major sources were accounted for and that changes to suspended sediments during transport (i.e., particle size, shape, composition) were minimal (Walden et al., 1997). A nonparametric Kruskal–Wallis test was performed on signatures passing all previous analyses to evaluate the presence of inter-category contrasts among the source samples (Collins and Walling, 2002; Barthod et al., 2015). Signatures showing no inter-category contrasts ($P > 0.05$) were eliminated from the dataset. Lastly, a stepwise discriminant function analysis was utilized to evaluate the discrimination power of each signature by determining the number of samples correctly classified (Barthod et al., 2015). This analysis also determined the best combination of signatures capable of discriminating source groups without redundancy based on the Wilk's lambda criterion. This optimum combination of fingerprints was used in the mixing model, MixSIAR to provide quantitative estimates of potential sources to suspended sediments in BBW (Mukundan et al., 2012). A more detailed description of the statistical selection of fingerprinting signatures and use of MixSIAR can be found in Chapter 2.2.5 and 2.2.6 respectively.

3.2.8 Application of spatial frameworks

Mixing models and their use in fingerprinting studies have increased rapidly in sophistication. Current models, such as MixSIAR, have user-friendly interfaces, and include hierarchical variance structure, incorporate complexities such as variability in fingerprints, discrimination factors, and concentration dependence (Phillips et al., 2014). For appropriate implementation of mixing models, "best practices" or guidelines for sensible use of these models is essential to maximize the models' usefulness but researchers should also be cognizant of their limitations and assumptions (Phillips et al., 2014). This is especially true when using stable isotope mixing models which were initially designed for food-web studies for sediment fingerprinting studies. Therefore in order to produce dependable results through MixSIAR, spatial frameworks as the collection of data and the incorporation of data into MixSIAR spreadsheets must be done with the utmost care to effectively characterize signature variability of sediments especially with regards to spatio-temporal scales.

For the sediment fingerprinting study in BBW, spatial frameworks were applied by strategically selecting suspended sediment samples and sediment source samples for use in uniquely designed spreadsheets capable of differentiating the various spatial frameworks in MixSIAR. The layout and unique design of each spreadsheet for use in MixSIAR was based on Stock and Semmens (2013) MixSIAR GUI User Manual and the various spreadsheet layouts provided within. Because spreadsheet examples provided by Stock and Semmens (2013) are intended for food-web research, it was necessary to compare predator-prey relationships given in the Stock and Semmens (2013) spreadsheets to the fingerprinting sediment and sediment source data. Therefore when comparing the food-web research examples used to describe the MixSIAR spreadsheets in Stock and Semmens (2013) to this sediment fingerprinting study, predator data was comparable to suspended sediment data and prey data was comparable to sediment source data. These comparisons were crucial in properly setting up the MixSIAR spreadsheets to reflect the various fingerprinting spatial frameworks in order to ensure each spatial framework was differentiated in MixSIAR.

Both Single Watershed Source Sampling and Regional Source Sampling under the SFS and Reach Source Sampling under the CFS used the same spreadsheet design, named the Geese Example in Stock and Semmens (2013). This spreadsheet design is unique in its inclusion of only one categorical covariate (i.e., group). For these spatial frameworks, the categorical covariate was represented by year. The specific design for the three separate spreadsheets (i.e., mixture, source and discrimination) which is required to run MixSIAR can be found by referring to the Geese Example in Stock and Semmens (2013). Appendix C, Appendix D and Appendix E contain the spreadsheets used for Regional Source Sampling, Watershed Source Sampling and Reach Source Sampling respectively. Multiple Watershed Source Sampling (i.e., SFS) and

Upstream Source Sampling (i.e., CFS) used a spreadsheet design modelled after the Wolf Example in Stock and Semmens (2013). This spreadsheet design includes multiple categorical covariates represented by "region" and "pack" in the Wolf Example and replaced by "year" and "site" for sediment fingerprinting purposes. The specific design for the three separate spreadsheets (i.e., mixture, source and discrimination) can be found by referring to the Wolf Example in Stock and Semmens (2013). Appendix F and Appendix G contain the spreadsheets used for Multiple Watershed Source Sampling and Upstream Source Sampling respectively.

3.3 Results

3.3.1 Selection of sources and signatures

Canonical discriminant function analysis results were identical amongst all spatial frameworks to the results presented in Chapter 2 (Fig 2.3.1) except for Reach Source Sampling. Canonical results for Reach Source Sampling were different in comparison to the canonical results presented in Chapter 2 because of sources to local (i.e., cultivated topsoil and streambank) and non-local sources (i.e., upstream suspended sediments) as a potential source (Figs. 3.7, 3.8, 3.9)

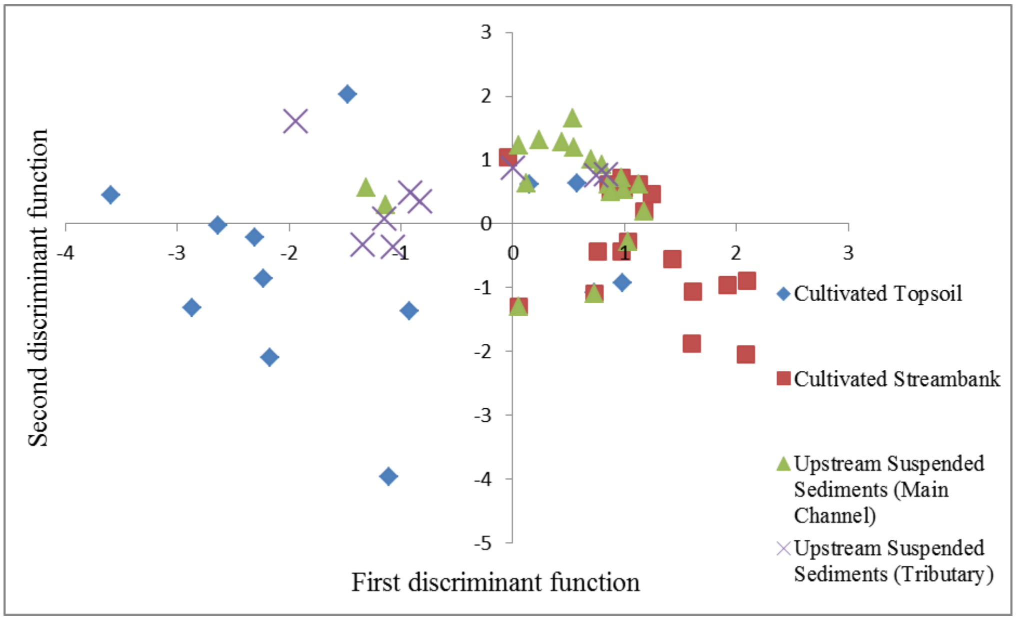

Figure 3.7 Canonical discriminant function analysis scores for Reach Source Sampling at the main outlet (SW1) of BBW for the following source types: 1) cultivated topsoil, 2) cultivated streambank, and 3) upstream suspended sediment from SW2 and SW8.

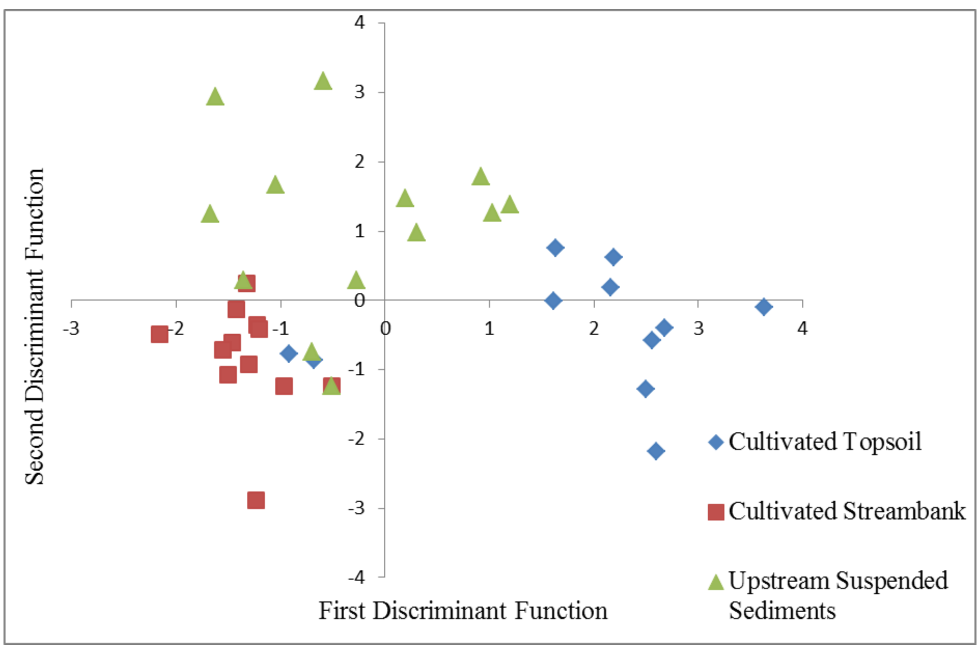

Figure 3.8 Canonical discriminant function analysis scores for Reach Source Sampling at SW2 in the BBW for the following source types: 1) cultivated topsoil, 2) cultivated streambank, and 3) upstream suspended sediments from SW3.

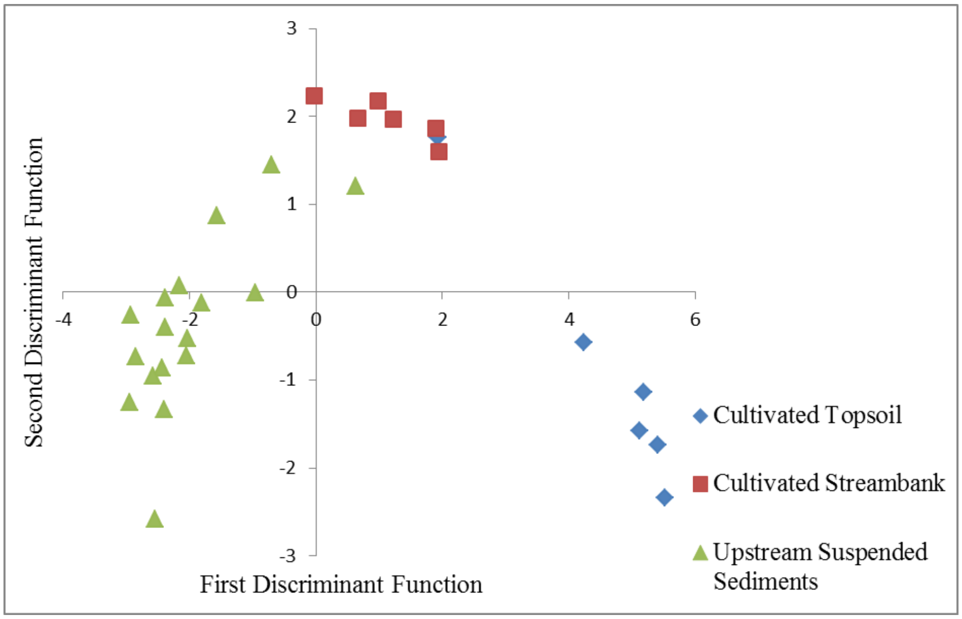

Figure 3.9 Canonical discriminant function analysis scores for Reach Source Sampling at SW3 in the BBW for the following source types: 1) cultivated topsoil, 2) cultivated streambank, and 3) upstream suspended sediments from SW4.

The plotted canonical scores for sediment sources utilized for Reach Source Sampling in BBW suggest that these sources were relatively easily distinguishable and could be used for source proportion modelling in MixSIAR.

Results from the Shapiro-Wilk normality tests, biplot analyses and Kruskal-Wallis tests for the selection of signatures for use in the stepwise discriminant function analysis produced identical results for Reach Source Sampling as the results obtained for Single Watershed Source Sampling in Chapter 2 (see sections. 2.3.2, 2.3.3, 2.3.4) for the selection of fingerprints intended for the stepwise discriminant function analysis. However, results obtained from the stepwise discriminant function analysis produced a different optimum combination of signatures for Reach Source Sampling compared to the optimum combination obtained in Chapter 2. Table 3.3 shows the selected fingerprints and the percentage of source samples correctly identified by using the optimum combination of fingerprints for sources contributing to suspended sediments at the main outlet of BBW using Reach Source Sampling.

Table 3.3 Results of the stepwise discriminant function analysis for Reach Source Sampling used to select the final combination of fingerprint properties essential for maximum source discrimination for potential source material: 1) cultivated topsoil, 2) cultivated streambank, 3) upstream suspended sediment from main stem of BBW, and 4) upstream suspended sediment from tributary

		Percentage of samples correctly classified				
Fingerprint Property	Fingerprint Combination	Total (%)	Cultivated Topsoil (%)	Cultivated Streambank (%)	Upstream W2 (%)	Upstream W8 (%)
V	V	51.1	61.5	91.9	0	5.6
R	V+R	56	38.5	89.2	0	50
L	V+R+L	64.3	69.2	86.5	25	50
U	V+R+L+U	70.2	61.5	89.2	31.3	72.2
B	V+R+L+U+B	70.2	61.5	86.5	43.8	66.7
Cs-137	V+R+L+U+B+Cs-137	70.2	69.2	89.2	31.3	66.7

Six variables including V, R, L, U, B, and Cs-137 were identified as being the optimum combination for discriminating sources in BBW using Reach Source Sampling. The optimal combination of signatures for sources contributing to suspended sediments at the main outlet of BBW produced an overall correctly classified percentage of 70 % (Table 3.3); the associated correctly classified source type percentages were as follows: 69 % for cultivated topsoil, 89 % for cultivated streambanks, 31 % for upstream suspended sediments from the main stem of BBW (i.e., SW2) and 67 % for upstream suspended sediments from a tributary of BBW (i.e., SW8). Results from the stepwise discriminant function analysis produced the same optimum combination of signatures for SW2 and SW3 as was produced for the main outlet of BBW (i.e., SW1) therefore the same suite of signatures were used for sediment apportionment using Reach Source Sampling (i.e., SW1, SW2, and SW3).

3.3.2 Simple Fingerprinting Study (SFS)

3.3.2.1 Single Watershed Source Sampling

Results for a SFS utilizing Single Watershed Source Sampling including the uncertainty assessment are included in Fig. 3.10

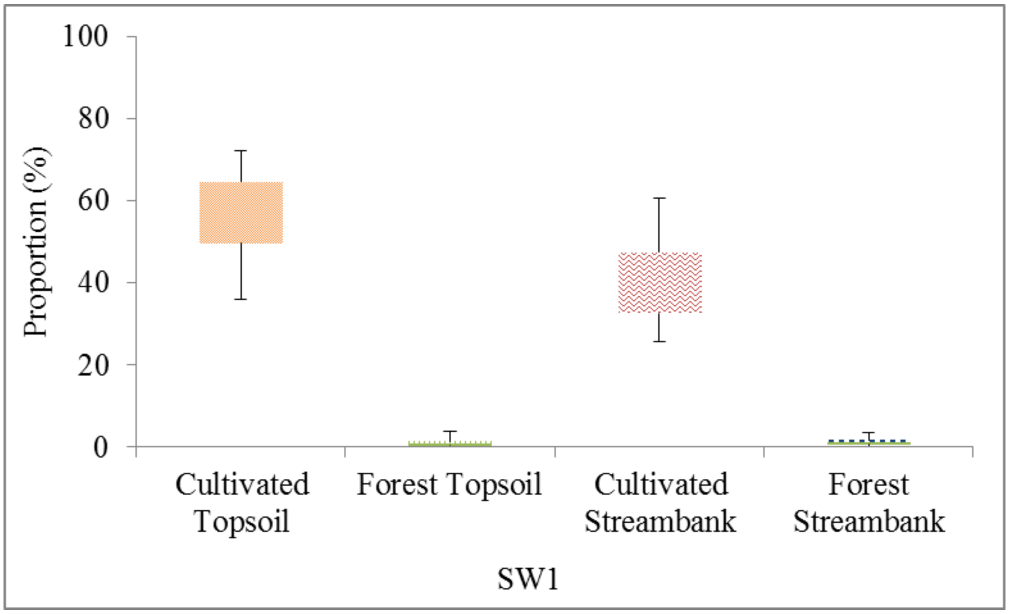

Figure 3.10 Box and whisker plot showing sediment source apportionment for SW1 using the following sources: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambank, and 4) forest streambank using Single Watershed Source Sampling.

At SW1, cultivated topsoil was found to be the predominant source of sediment, accounting for approximately 50 to 65 %. The model identified cultivated streambank as accounting for approximately 33 to 47 %. Forest streambanks were estimated to account for approximately 1 to 2 %. Forest topsoil was found to contribute the smallest amount of sediments, accounting for approximately 0 to 2 %.

3.3.2.2 Regional Source Sampling

Results for a SFS utilizing Regional Source Sampling to determine source apportionment results for sediments collected at the main outlet of BBW are presented in Fig. 3.11.

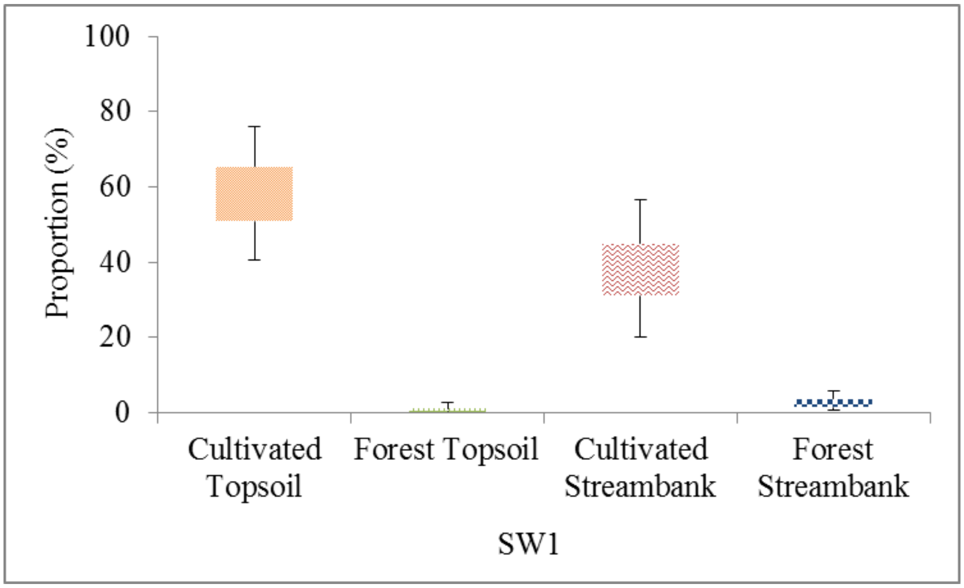

Figure 3.11 Box and whisker plot showing sediment source apportionment for SW1 using the following sources: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambank, and 4) forest streambank for Regional Source Sampling where source samples were collected from both BBW and Little River watershed.

Results show the influence of using source samples collected outside the watershed boundary as additional potential sources on estimates of source apportionment. For sediments collected at the main outlet of BBW, cultivated topsoil contributed 51 to 65 %, cultivated streambanks contributed 31 to 45 %, forest streambanks contributed 2 to 4 % and forest topsoil were found to contribute the least at 0 to 1 %. These source proportion estimates vary slightly from estimates made by a Single Watershed Source Sampling, suggesting that adding additional source samples had little effect on the final source estimates. Although Regional Source Sampling had little influence in the case of BBW, this does not suggest that all watersheds will see minimal influence on sediment apportionment estimates. It is likely that by utilizing more source samples than what was used in BBW (i.e., 3 forest topsoil and 10 forest streambanks samples) an influences may be observed on apportionment. It is also important to note that Regional Source Sampling is not restricted to a single target location, it may also be applied to multiple target locations. For simplicity, however, only results for a SFS focusing on the main outlet of BBW are depicted in the current study. This spatial framework may be used in any watershed requiring additional samples, with conditions making sampling outside watershed boundaries more practical.

3.3.2.3 Multiple Watershed Source Sampling

Simple Fingerprinting Study utilizing Multiple Watershed Source Sampling for multiple locations of interest (SW1, SW2, SW3, SW4, and SW8) are presented in Fig. 3.12.

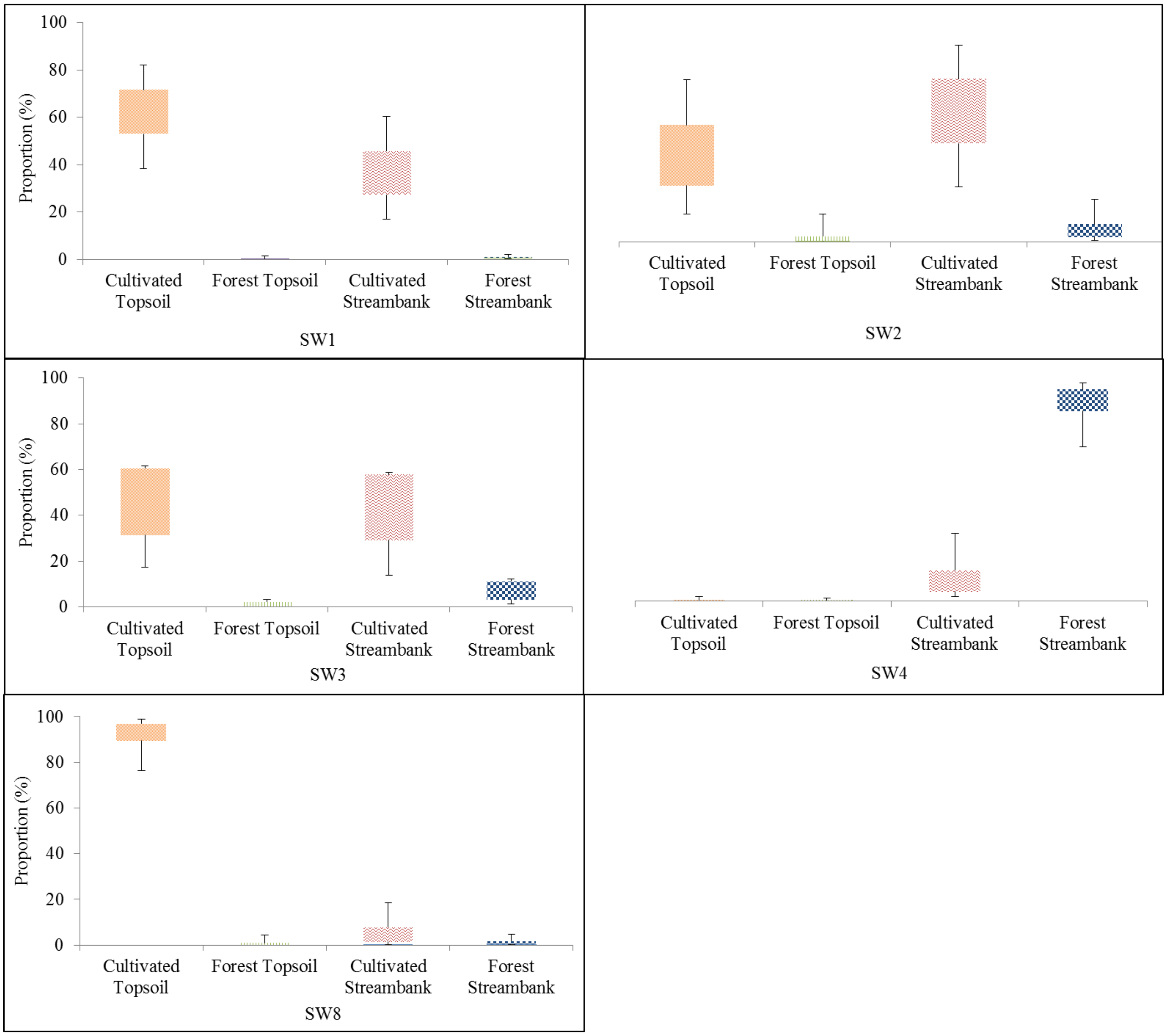

Figure 3.12 Box and whisker plots showing sediment source apportionment for SW1, SW2, SW3, SW4, and SW8 using the following sources: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambanks, and 4) forest streambanks for Multiple Watershed Source Sampling. .

For suspended sediments collected at the main outlet of BBW: cultivated topsoil was estimated to contribute 53 to 72 %, cultivated streambanks contributed 27 to 46 %, forest streambanks contributed 0 to 1 % and forest topsoil contributed 0 %. Similarly, sources of sediments collected at SW3 and SW8 also concluded the same sequence of predominant sources, with sediment source contributions at SW3 being as follows: cultivated topsoil contributed 31 to 61 %, cultivated streambank contributed 29 to 58 %, forest streambank contributed 3 to 11 % and forest topsoil contributed 0 % and source contributions at SW8 were as follows: cultivated topsoil contributed 89 to 97 %, cultivated streambank contributed 2 to 8 %, forest streambank contributed 0 to 2 % and forest topsoil contributed 0 to 1 %. Sub-watershed 2 and SW4 differed in sediment apportionment with source proportion estimates at the outlet of SW2 predominantly originating from cultivated streambank (41 to 69 %), cultivated topsoil (24 to 49%), forest streambanks (2 to 7 %) and forest topsoil (0 to 2 %) and sediment collected at the outlet of SW4 originating predominantly from forest streambank (85 to 95 %), cultivated streambank (4 to 14 %), forest topsoil (0 to 1 %) and lastly cultivated topsoil (0 to 1 %).

These results correspond well with the landuse in BBW due to SW1, SW3 and SW8 consisting of predominantly agricultural land with 77 %, 77 % and 66 % agricultural landuse respectively based on local catchment areas (Tables 3.1 and 3.2). SW2 is composed of the following landuse: 55 % agriculture, 36 % forest and 9 % other; because it has significantly less agriculture than SW1, SW3 and SW8 this may provide reasoning for the difference in the leading source of suspended sediments at SW2. SW4 had the greatest variation in sediment source contributions with findings suggesting the majority of sediments are originating from forest streambank sources. The landuse in SW4 consists of 65 % forest, 21 % other (i.e., wetlands and residential) and 14 % agriculture (Table 3.1) which corresponds well to the sediment source proportion estimates, suggesting the importance of landuse on sources of suspended sediments in BBW.

3.3.3 Complex Fingerprinting Study (CFS)

3.3.3.1 Upstream Source Sampling

Sediment source apportionment results for a CFS using Upstream Source Sampling are depicted in Fig 3.13. At the main outlet of BBW (i.e., SW1) and SW3 and SW8, cultivated topsoil, cultivated streambanks, forest streambanks and forest topsoil were determined to be the main sources of sediments in descending order. For sediments collected at the main outlet of BBW, cultivated topsoil contributed 51 to 69 %, cultivated streambanks 30 to 48 %, forest streambanks 0 to 1 % and forest topsoil 0 %. For SW3, cultivated topsoil contributed 32 to 57 %, cultivated streambank 33 to 59 %, forest streambanks 3 to 10 % and forest topsoil 0 to 2 %. For SW8, cultivated topsoil contributed 80 to 96 %, cultivated streambanks 2 to 13 %, forest streambanks 0 to 1 % and forest topsoil 0 to 1 %.

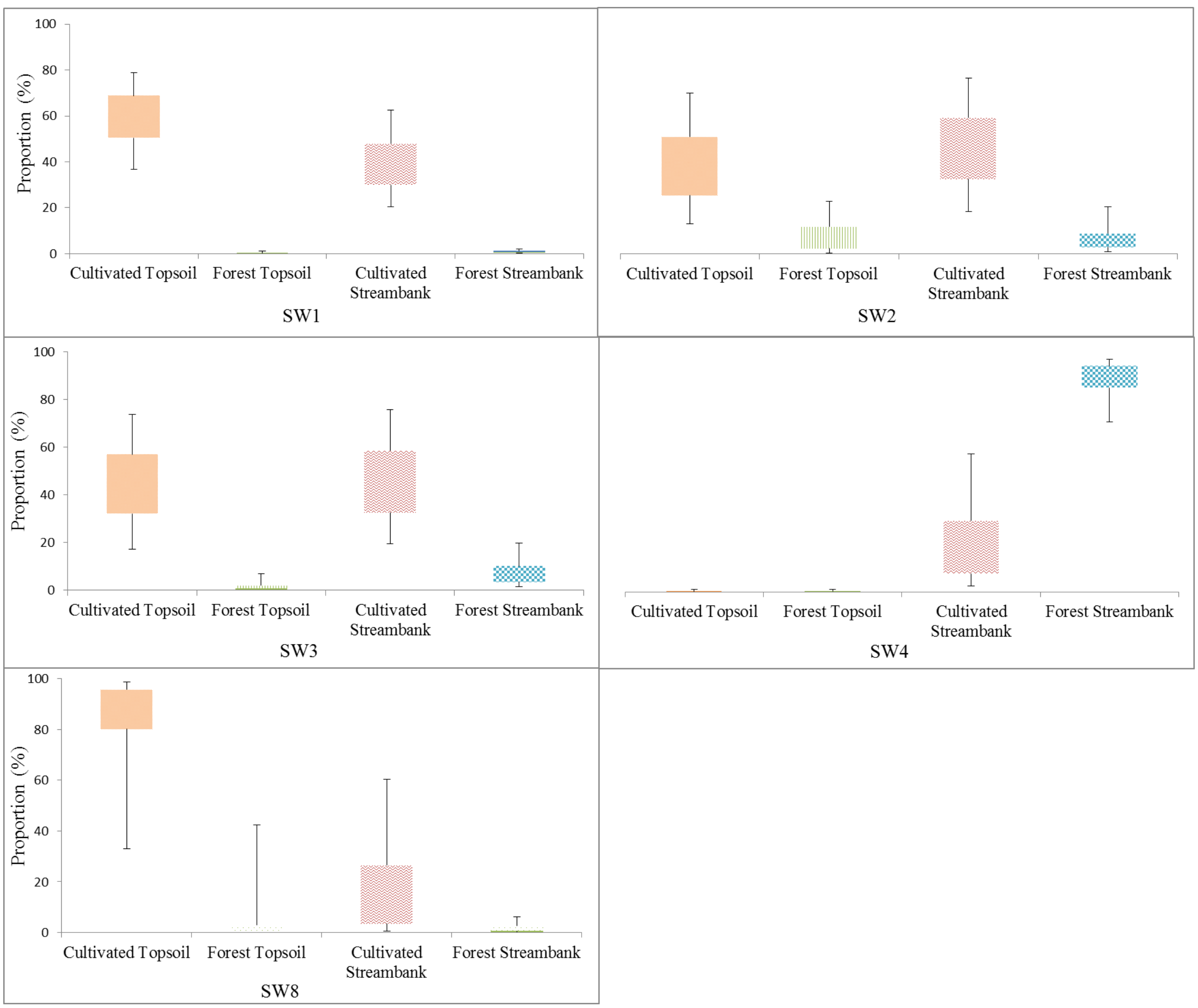

Figure 3.13 Box and whisker plots showing sediment source apportionment for SW1, SW2, SW3, SW4, SW8 using the following sources: 1) cultivated topsoil, 2) forest topsoil, 3) cultivated streambanks, and 4) forest streambanks for Upstream Source Sampling.

Suspended sediment samples collected at the outlet of SW2 differed slightly in source proportion estimates with the predominant source of sediments being cultivated streambank (32 to 59 %), cultivated topsoil (25 to 50 %), forest streambank (3 to 9 %), and forest topsoil (3 to 12 %). Sub-watershed 4 had the greatest variation in sediment source contributions with 85 to 94 % from forest streambanks, 5 to 14 % from cultivated streambanks, 0 to 1 % from forest topsoil and 0 % from cultivated topsoil.

These results follow the same pattern as the results obtained for Watershed Source Sampling and also correspond well to landuse in BBW, with SW1 (main outlet of BBW), SW3 and SW8 consisting of the greatest agricultural landuse and results suggesting the same sequence of predominant sediment sources between the three target locations. This indicates the influence that intensive cultivation has on the creation of sediments within BBW. With a slightly lower agricultural landuse, source proportion estimates for SW2 found that erosion of cultivated streambanks were the leading cause of suspended sediments. Furthermore, the switch in landuse from predominantly cultivated in the SW1, SW2, SW3, and SW8 to predominantly forest in SW4 also changed the source proportion estimate results which suggested that the greatest contributor of suspended sediments in SW4 was forest streambanks. These results correspond to the findings of a SFS employing Multiple Watershed Source Sampling but contain larger error bars suggesting that more uncertainty is associated with Upstream Source Sampling. This was expected due to Upstream Source Sampling only considering source samples collected upstream as potential sources of sediments, thereby reducing the number of samples representing each source type.

3.3.3.2 Reach Source Sampling

Sediment source apportionment results obtained using Reach Source Sampling are presented in Fig. 3.14. Reach Source Sampling was applied to SW1, SW2 and SW3 in BBW. For the main outlet of BBW the predominant sources of suspended sediments in descending order were: non-local from the main stem of BBW (32 to 43 %), local cultivated topsoil sources (27 to 37 %), local cultivated streambank sources (14 to 22 %) and non-local sources from a tributary of BBW (8 to 15 %). Local contributions were estimated to contribute 41 to 59 % and non-localized sources were estimated to contribute slightly less at 40 to 58 %.

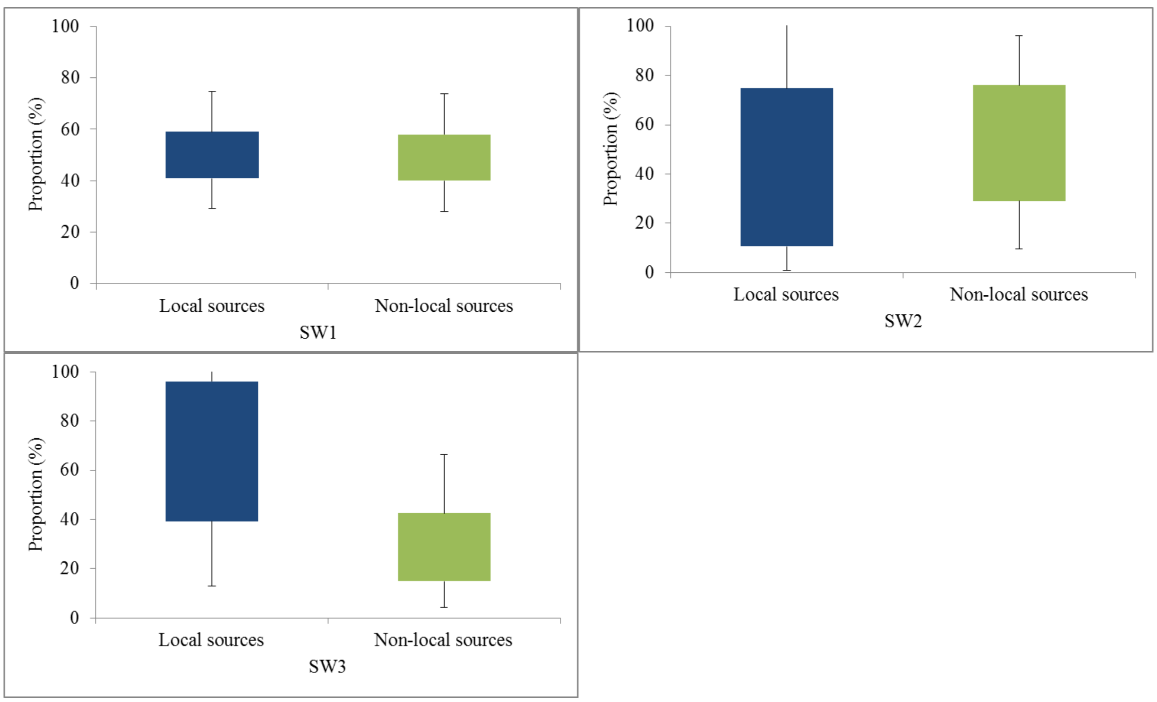

Figure 3.14 Box and whisker plot showing sediment source apportionment for SW1, SW2, and SW3 using the following sources: 1) local, and 2) non-local source for Reach Source Sampling.

For SW2, contributions to suspended sediments were as follows: non-local sources from the main stem of BBW (29 to 76 %), local cultivated streambank sources (6 to 38 %) and local cultivated topsoil sources (5 to 37 %). Source proportion estimates suggest that non-local sources (29 to 76 %) were a greater source of suspended sediments then in comparison to local sources

(11 to 75 %). For SW3, predominant sources of suspended sediments were: local cultivated topsoil sources (31 to 63 %), non-local sources from the main stem of BBW (15 to 43 %) and local cultivated streambank sources (9 to 34 %). Therefore, for SW3 source proportion estimates suggest that local contributions (40 to 97 %) were greater than non-local sources (15 to 43 %). Results seem to suggest that for sub-watersheds containing high agricultural landuse, including SW1 (77 % agriculture) and SW3 (77 % agriculture) local source tend to be a greater contributor to suspended sediments then sub-catchments such as SW2 (55 % agriculture) where the agricultural landuse is less.

3.4 Discussion

3.4.1 Sediment source apportionment variability among spatial frameworks

Results for the sediment source apportionment estimates between the two different watershed studies (i.e., SFS and CFS) and their associated source sampling frameworks are presented in Table 3.4.

Table 3.4 Sediment source apportionment results for different spatial frameworks for sediments collected at the main outlet of BBW

			Sediment Source Apportionment				
Fingerprint Study	Source Sampling Framework	Total No. Source Samples	Cultivated Topsoil (%)	Forest Topsoil (%)	Cultivated Streambank (%)	Forest Streambank (%)	Non-local Sources (%)
Simple	Regional	149	51 - 65	0 - 1	31 - 45	2 - 4	---
	Single Watershed	136	50 - 65	0 - 2	33 - 47	1 - 2	---
	Multiple Watershed	136	53 - 72	0	27 - 46	0 - 1	---
Complex	Upstream	136	51 - 69	0	30 - 48	0 - 1	---
	Reach	84	27 - 37	---	14 - 22	---	40 - 48

In comparison to the preceding study (Chapter 2), which utilized Single Watershed Source Sampling to determine sediment source apportionment for the main outlet of BBW (i.e., SW1), it was expected that many of the other spatial frameworks (i.e., Multiple Watershed Sampling, Upstream Source Sampling) would produce the same apportionment estimates for SW1, due to the source sample inventory being the same. However, the apportionment estimates were not the same between these spatial frameworks, which may have been attributed to the use of the MixSIAR, MCMC run length of “medium” over the recommended MCMC run length of “very long”. The use of the “medium” run length likely resulted in the discrepancies between sediment source apportionment estimates in Table 3.4 between Single/Multiple Watershed Source Sampling and Upstream Source Sampling. A SFS utilizing Regional Source Sampling produced sediment source apportionment results that were the most similar to the results presented in Chapter 2 (i.e., SFS utilizing Single Watershed Source Sampling). Source proportion estimates were almost identical between the two spatial frameworks indicating that the addition of source samples from Little River watershed had little influence on source proportion estimates.

Source proportion estimates were also particularly similar for all sediment sampling locations included in SFS utilizing Multiple Watershed Source Sampling and a CFS utilizing Upstream Source Sampling. Both Multiple Watershed Source Sampling and Upstream Source Sampling concluded the same sequence of predominant sediment sources for SW1, SW3, SW4 and SW8, while SW2 varied slightly with regards to the sequence of predominant sources. Reach Source Sampling provided a different way of interpreting source proportion estimates compared to the other spatial frameworks. Unlike the other spatial frameworks, Reach Source Sampling is capable of quantifying the proportion of sources coming from non-local sources against more local sources. Source proportion estimates for the overall contribution for local versus non-local sources suggest that at the main outlet of BBW and SW3, the main source of suspended sediments were local sources while non-local sources were the greatest contributor of suspended sediments in SW3. These results suggest that sub-watersheds containing a higher agricultural landuse tend to have high local source contributions compared to sub-watershed with less agricultural landuse. These results suggest the influence of landuse on sediment origin, therefore in order to effectively manage and control suspended sediment the landuse within watersheds of interest must be known.

3.4.2 Spatial specificity and uncertainty

The box and whisker plots provided in the previous section (section 3.3) give information on the uncertainty of the data associated with the model results. By observing the distance between the first and third quartiles it is possible to assess the uncertainty which may be present in the source proportion estimates. Results for the SFS using a single target location (the main outlet) suggest that the greatest amount of error was associated with cultivated topsoil followed closely by cultivated streambanks, with minimal error associated with forest topsoil and forest streambank sources likely due to the minimal contribution from the two sources. Similarly, uncertainty assessments for Regional Source Sampling results yielded the same conclusion as determined for Single Watershed Source Sampling.

Results for a SFS using Multiple Watershed Source Sampling suggest that the greatest amount of uncertainty was associated with cultivated topsoil and cultivated streambank sources with the smallest amount of uncertainty associated with forest topsoil and forest streambank sources. Similar to Single Watershed Source Sampling and Regional Source Sampling, the low uncertainty associated with forest sources are likely due to the minimal contributions from forested sources. Similarly, Upstream Source Sampling results suggest substantial uncertainty with cultivated topsoil and cultivated streambank sources, with minimal uncertainty associated with forest topsoil and forest streambank sources, except for SW8 where forest topsoil contained large error bars suggesting that forest topsoil sources may in fact contain a great deal of uncertainty but its relatively small sediment source contributions allows for its uncertainty assessments to be perceived as minimal. Lastly, Reach Source Sampling produced results that contained high uncertainty. This is likely due to non-local sources being composed of a mixture of soils eroded from many different landuse sources causing highly variable data.

A difference in the extent of uncertainty was also observed between different spatial frameworks. Relative to the SFS employing Single Watershed Source Sampling, Regional Source Sampling contained slightly different spatial specificity due to some of the samples being collected from outside watershed boundaries; however, overall uncertainty associated with sediment source types was nearly identical for both spatial designs. These two spatial frameworks were not able to detect a switch in the predominant sources of sediments from the headwaters to the outlet creating considerable uncertainty with regards to the true sediment dynamics within the watershed. However, they were able to provide an overall assessment of contributions from different sources within the watershed. Alternatively, spatial frameworks with a great degree of spatial specificity, such as a CFS employing Reach Source Sampling, contained a lower number of samples, increasing the overall uncertainty associated with sediment source types relative to the conventional method. However, Reach Source Sampling was able to detect changes in sediment source contributions along the length of Black Brook which the spatial frameworks containing little spatial specificity (i.e. SFS Single Watershed Source Sampling and Regional Source Sampling) were unable to detect. Nested spatial frameworks such as a SFS employing Multiple Watershed Source Sampling and a CFS employing Upstream Source Sampling contained a great degree of spatial specificity; however source sample numbers for the main outlet were similar to the spatial frameworks with low spatial specificity (Table 3.5) and therefore uncertainty associated with sediment source types were kept relatively low relative to the more conventional results presented in Chapter 2. Furthermore, these spatial frameworks were also able to detect the switch in sediment sources from the headwaters to the outlet of BBW due to the nested design of these spatial frameworks increasing the dependability of nested spatial frameworks.

3.4.3 Capturing sediment dynamic at a watershed scale

Results obtained in the current chapter show the variability associated with different spatial frameworks as well as the extent sediment dynamics within the entire area of BBW are represented by each spatial framework. Many of the spatial frameworks presented in this review including Multiple Watershed Source Sampling and Upstream Source Sampling are capable of achieving precision soil conservation strategies while other spatial frameworks such as Single Watershed Source Sampling and Regional and Reach Source Sampling would not be capable of precision soil conservation strategies. Spatial framework is extremely important in watersheds, such as BBW, which are heterogeneous with regards to their geology, pedology and landuse, thereby resulting in signatures often providing lower source discrimination if spatial frameworks are not designed to capture these heterogeneous features. The large variability of environmental variables over spatial and temporal scales, the source, fate, and transport processes are difficult to predict and model precisely making it difficult, especially in large catchments with varying landuse, to accurately estimate source contributions (Davis and Fox, 2009). Spatial and temporal variation may decrease the efficiency of the fingerprinting procedure if the spatial framework is not designed to meet the specific requirements of the watershed. In BBW where landuse changes considerably from the headwaters to the outlet, collecting suspended sediments solely at the main outlet is likely inadequate in capturing spatial variability in the origin of sediments and implementing one type of BMP is likely inadequate at effectively combating against sediments. Results suggest that landuse has a substantial influence on suspended sediment source contributions in BBW due to source apportionment results changing along the length of Black Brook as landuse changes. Near the outlet where there is predominantly agriculture and high-order streams, the predominant source of sediments according to the nested spatial frameworks were cultivated sources while in the headwaters, where there are predominantly forests and low-order streams, the predominant source of sediments were forest sources. These finding follow conclusions made by Koiter et al. (2013b) and Barthod et al. (2015) which found a switch in the predominant sediment sources between the headwaters and the outlet, highlighting the need for multiple nested spatial frameworks and emphasizes the importance of precision soil conservation strategies.

Many fingerprinting studies have noted that only small proportions of eroded sediments within river basins make it to the outlet (Koiter et al., 2015). This is often attributed to the net storage of sediment created by the transport of suspended sediments downstream towards the outlet (Koiter et al., 2013b). Sediment delivery ratios typically decrease with increasing drainage area as there is a decline in the connectivity and an increase in the residence times and sediment storage (Koiter et al,. 2013b; Walling, 1983; Fryirs, 2013). In BBW many physiographic features including the change from low-order streams in the headwaters to a high-order streams near the outlet, along with the presence of beaver dams and many weirs, vegetative barriers and floodplains emphasize the importance of multiple, carefully selected suspended sediment sampling locations along the length of Black Brook. Therefore, in BBW, solely sampling at the watershed outlet is likely to give an incomplete assessment of sediment sources because of the incomplete representation of the processes affecting sediment dynamics (Koiter et al., 2013b).

Furthermore because BBW is composed of several sub-watersheds which according to Brosinsky et al. (2014) creates considerable variability in source apportionment, emphasis is needed for comparisons between each sub-watershed in order to assess the variability in sediment source proportions. Furthermore, sampling solely at the main outlet does not capture variations in source contributions created from variations in landuse across the watershed, specifically with regards to the sediments generated as landuse changes from the headwaters to the outlet.

Therefore, it is very important to explore spatial framework options due to each framework having its merits and ability to tell one part of the story with regards to sediment dynamics at a watershed scale. It may be necessary to do multiple analyses to have a more complete understanding of the system in order to fully represent the true origin of sediments at a watershed scale. However, it is important to note that only nested spatial frameworks were capable of detecting a switch in the sources of sediments from the headwaters to the outlet of the watershed. However, the results obtained from SFS utilizing Multiple Watershed Source Sampling and a CFS utilizing Upstream Source Sampling produced source proportion estimates that best matched the objectives of this study and in our opinion corresponded best to field observations made within the watershed. Source proportion results from Multiple Watershed Source Sampling and Upstream Source Sampling contained less uncertainty compared to the results from Reach Source Sampling and therefore it is suggested that either of these spatial frameworks is used to support of precision soil conservation strategies for BMP implementation in BBW.

3.5 Conclusions

The current study demonstrated the variation in source proportion estimates and their relative uncertainty estimates between spatial frameworks. In summary, the results of the sediment fingerprinting study in the BBW based on 7 years (2008-2014) of sediment sampling, demonstrated that there was a switch in suspended sediment sources between the headwaters and the outlet of the watershed. Only nested spatial frameworks, composed of multiple sampling locations found along the length of BBW, were capable of detecting this switch in sediment sources with sediments in the upper reaches of BBW originating from predominantly forest sources and sediments in the lower reaches originating from predominantly cultivated sources. The results of the current chapter highlight the importance of spatial frameworks. The main question of the current chapter was whether a single sediment sampling location installed at the outlet of BBW was providing a complete picture of sediment origins. The current study demonstrated that in BBW, a single suspended sediment sampling location was not adequate in assessing sediment source contributions and would likely lead to poor management decisions. In the case of sediment provenance in BBW, a SFS utilizing a single sediment sampling location would have given an incomplete assessment of the sources of sediments due to sediments collected at the main outlet not fully representing processes affecting sediment dynamics elsewhere in the watershed especially the headwaters where landuse is much different.

To meet our final objective, the current study recommends the use of nested spatial frameworks in BBW, due to the heterogeneity of the watershed resulting in the nested spatial framework corresponding best to the difference in landuse, field observations and results stemming from previous studies conducted in the watershed. Nested spatial frameworks were capable of providing a comprehensive understanding of the sediments being transported within the watershed through strategic and thorough monitoring, capable of producing detailed information on spatial variably as well as the connectivity of suspended sediment (Carter et al., 2006). Above all, this study proved the importance of understanding the strategic placement of sampling locations as it will provide more valuable information on sediment dynamics at a watershed scale, leading to more meaningful management decisions.

3.6 References

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4. CONCLUSIONS AND FINGERPRINTING PERSPECTIVES

This study identified sources of the suspended sediments in the Potato Belt of New Brunswick in Atlantic Canada based on 7 years (2008 – 2014) of sediment sampling and contributed to the overall sediment fingerprinting literature by providing contrasting spatial frameworks for future studies intending to implement sediment fingerprinting within other watersheds. The results of this research are the first of its kind in Atlantic Canada and have helped to enhance the understanding of sediment sources and dynamics in the intensively cultivated Black Brook Watershed (BBW), which has been identified as being prone to some of the most severe water erosion in Canada.

The initial study (i.e., Chapter 2) has contributed to the overall project by providing sediment source proportion estimates for BBW using a SFS framework, which considers each watershed as a single catchment and entails the collection of suspended sediments at a single sampling location (e.g., the main outlet) and allows for watershed scale conclusions based on these samples (i.e., Single Watershed Source Sampling). A SFS, utilizing Single Watershed Source Sampling, concluded that the main source of suspended sediments within the watershed were predominantly cultivated topsoil (50 to 65 %), followed by cultivated streambank (33 to 47 %), forest streambank (1 to 2 %) and forest topsoil sources (0 to 2 %). Results were consistent with our hypotheses that upland erosion was the dominant source of suspended sediments to Black Brook and the landuse percentages of 65 % agriculture, 21 % forested and 14 % residential or wetland in the watershed. It was concluded that sampling solely at the main outlet of the watershed may incompletely represent sediment dynamics at a watershed scale, therefore, there is ultimately a need to explore more robust spatial frameworks in order to determine whether different spatial frameworks are better able to describe sediment dynamics at a watershed scale.

The second study, described in Chapter 3, presented spatial frameworks intended to expand the sediment fingerprinting study in BBW by providing in some cases, a more complete assessment of sediment dynamics through more robust spatial frameworks. Spatial frameworks presented in Chapter 3 were divided into two groups: 1) SFS, and 2) CFS, with the main difference between the two strategies being the treatment of a watershed as either: a) single homogeneous catchment (i.e., SFS) or b) many smaller nested heterogeneous sub-watersheds (i.e., CFS). The results presented in Chapter 3 demonstrate the significance of spatial frameworks on sediment source apportionment estimates. Simple Fingerprinting Studies utilizing Single Watershed Source Sampling and Regional Source Sampling were incapable of detecting spatial variations in sediments along the length of Black Brook; however SFS and CFS containing a nested framework (i.e., Multiple Watershed Source Sampling, Upstream Source Sampling and Reach Source Sampling) were able to detect spatial variations in sediment origins.

Further information on sediment origin was gathered from a CFS utilizing Reach Source Sampling, as this framework provided information on contribution of local sediment sources compared to non-local sources, confirming that for many target locations along Black Brook the majority of sediments originated from non-local sources. Reach Source Sampling results however, did contain the largest error bars relative to the alternative frameworks meaning that the source proportion results contained the greatest amount of uncertainty. Upstream Source Sampling also contained large error bars and therefore contained a great amount of uncertainty as well. However Reach Source Sampling and Upstream Source Sampling were capable of completely representing sediment dynamics at a watershed scale. Conclusions on the switch in the sources of sediments between the headwaters and the watershed outlet would not have been realized by referring to merely the information provided in Chapter 2.

4.1 Implications for soil conservation in Atlantic Canada

Impacts of suspended sediments and agrochemicals on water quality and soil erosion on the reduction of soil fertility, loss of nutrients, and decline of crop yields are major environmental concerns facing the agricultural sector (Chow et al., 2011). The current study verified research done by Saini and Grant (1980) which reported soil losses of 17-40 t ha^{-1} yr^{-1} and research by Chow et al. (1990) which reported losses of 1.2 to 24.3 t ha^{-1} yr^{-1} using plot measurements which attributed soil losses to upland erosion due to shallow soils, rolling topography coupled with cultural practices including up-and-down slope cultivation, worsened by climatic conditions. Furthermore, previous sediment load estimates of 181.6 t km^{-2} y^{-1} which was estimated by Chow et al. (2011) can now be attribute to specific sediment source areas. This allowed discrepancies between field level erosion measurements (e.g., Saini and Grant, 1980; Chow et al., 1990; Chow et al., 1999) and stream monitoring of sediment loading (e.g., Chow et al., 2011) to be resolved.

Through the current study it was realized that although upland erosion was found to be the main contributor to suspended sediments in Black Brook, streambank erosion in agricultural areas is also a significant source of instream sediments. Streambank erosion in agricultural areas was found to be the second greatest contributor to suspended sediments along the length of Black Brook, according to most spatial framework outputs. Since its establishment as a benchmark watershed in 1992, the majority of research studies in BBW have focused predominantly on upland erosion from agriculture. Furthermore many of the Beneficial Management Practices (BMPs) that have been implemented within the watershed have been focused on preventing upland erosion as well. It is estimated that flow diversion terraces have been implemented on approximately 50 % of cultivated lands and 24 % of the total watershed area in BBW (Yang et al., 2009). The current study has shown that sediment fingerprinting has a practical implication on BMPs, because the methods used for upland erosion control are different from those used for streambank erosion control (Mukundan et al., 2010). Therefore, it is recommended that more consideration be given to BMPs that prevent streambank erosion in agricultural areas. Beneficial Management Practices such as wider riparian areas or bank stabilization efforts should be considered in the watershed according to the findings of this study.

The current study was able to demonstrate that sediment fingerprinting offers an additional method of aiding in the developing and targeting of BMPs especially with regards to precision soil conservation strategies . The sediment fingerprinting technique may be improved by using it in addition to field experiments (e.g., Chow et al., 1999) and/or utilizing integrated models such as SWAT (e.g., Yang et al., 2009). Sediment fingerprinting is specifically useful at identifying and quantifying sources of suspended sediments throughout the watershed in order to implement more effective BMPs to manage sediments. In the case of BBW, although cultivated topsoil was found to be the predominant sediment source contributing to suspended sediments, the watershed already has many measures put in place to combat against upland erosion and, therefore, it is recommended that additional measures are considered to combat against what was determined to be the second leading cause of suspended sediments, streambank erosion in agricultural areas. By implementing BMPs to target streambank erosion, the thick layer of silt that covers the streambed near harvest and is later flushed downstream in the spring may be reduced, benefiting those downstream that use this water as a drinking source.

The results of the current study follow a long line of other sediment fingerprinting studies (i.e., Koiter et al., 2013b; Gellis and Noe, 2013; Voli et al., 2013) with a similar finding, that streambank sources are a major source of instream suspended sediments than what was previously assumed. Although there are an assortment of conditions within BBW including: terrain that is undulating, with long and steep slopes, coarse to fine-textured soils that are easily erodible and land that is scarce that has led to intensive potato production with potato-grain and potato-potato-grain rotations, there are still additional measures that may be taken to reduce soil and water degradation in the area (Rees et al., 1999). Through the current sediment fingerprinting study, it is now clear where the majority of sediments are originating from and therefore more effective measure may be taken to improve soil conservation in Atlantic Canada.

4.2 Recommendations for future research

Future sediment fingerprinting studies are encouraged to confirm that fingerprinting results are complete and dependable. Results from the current study suggest that a robust spatial framework is a prerequisite to achieving complete fingerprinting source proportion estimates to aid in the selection of the most appropriate BMPs through precision soil conservation strategies (Mukundan et al., 2010). Each spatial framework has its merits and is capable of describing an important part of what is happening in a watershed with regards to sediment dynamics. For similar studies, it is recommended that multiple analyses are conducted to explore spatial framework options, similar to what was done in the current study, in order to obtain a more complete understanding of the watershed system. For example, although crude, the conventional SFS employing Single Watershed Source Sampling provides an overall assessment of sediment dynamics within the watershed. However, nested spatial frameworks are capable of describing more about the sediment dynamic in the sub-watersheds, but are less capable in some cases (i.e., Reach Source Sampling) at describing what is happing in the watershed as a whole. Therefore, future sediment fingerprinting studies are encouraged to explore alternative spatial framework options as was presented in Chapter 3 to accurately represent sediment dynamics. Future sediment fingerprinting researchers are encouraged to properly design their spatial framework strategy to ensure watershed heterogeneity is captured. Future sediment fingerprinting studies are encouraged to explore nested spatial frameworks over relying solely on conventional methods (i.e., SFS Single Watershed Source Sampling) as exploring different spatial framework options provides the researcher with a better understanding of the watershed of interest.

The current study would have benefited from adding additional source samples to increase the overall sample number to preferably 30 samples or more per source type in order to comment on the nature of the variance for the soil properties (Lobb, 2016). For example, the current study contained only 14 individual samples for forest topsoil sources and 9 individual samples for forest streambank sources which is likely insufficient to adequately characterize the soil properties within these two source types. As discussed previously, the estimate of 30 samples per source type is derived from a soil fertility study conducted by Slevinsky et al. (2001, 2002), due to there being no sediment fingerprinting study, to our knowledge, that have determined the optimum number of source samples required to acquire dependable sediment fingerprinting results. It is, therefore, recommended that future research explore the amount of source samples required to adequately comment on the mean and variance of soil properties. The amount of samples required will likely vary depending on the soil conditions (i.e., sampling in a potato field vs. forested area) however these sample number requirements for particular field conditions should be made aware for future research purposes.

This study also may have benefited from the use of additional composite signatures, such as geochemical elements, that respond very differently to environmental controls (Mukundan et al., 2012). This would have likely resulted in better discrimination between potential sources and would have provided more accurate estimates of sediment source apportionment (Mukundan et al., 2012). Sediment and sediment source samples collected in BBW were in fact analyzed for geochemical elements, however, due to a lack of sufficient time, no statistical analyses or modelling was completed on this geochemical data. Therefore, any future sediment fingerprinting work in BBW should be encouraged to use this geochemical data to provide better discrimination.

In conclusion, implementing a successful sediment fingerprinting study begins with the spatial framework, therefore it is recommended that researchers and managers intending to implement sediment fingerprint within a watershed ensure that adequate consideration is given to the spatial framework designed for the proposed area for sediment fingerprinting. Exploring different spatial frameworks by completing different analyses to obtain a better understanding of sediment dynamics within a watershed is crucial. The utility of sediment fingerprinting can be improved by carefully considering which locations to sample and recognizing the effect this choice will have on data interpretation. Secondly, ensuring that an adequate number of source samples are collected to adequately comment on the mean and variance of soil properties is crucial if dependable sediment fingerprinting source apportionment estimates are to be made. Thirdly, ensuring that composite fingerprints are used that respond very differently to environmental controls is recommended in order to provide more accurate estimates of sediment source apportionment.

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APPENDICES

Appendix A: Comparison between sample preparation methods for particle size and shape

The majority of instruments used to measure particle size and shape are composed of both a liquid and dry dispersing unit used to present the sample to the measurement zone. It is preferred that wet dispersing units are used for particle size and shape analysis because the transport of the subsample to the measurement zone mimics the fluvial transport within the original channel providing more accurate representation of how the particles would move based on their size and shape. Two methods exist for preparing suspended sediment samples for wet dispersing units used for particle size analysis. The first method consists of collecting suspended sediments from a channel and keeping the subsample in solution with water from the channel; these subsamples remain in solution from the moment they are collected in the channel to when they are analyzed. The second method consists of collecting suspended sediments from a channel and bringing the subsample to the laboratory, drying the subsample and subsequently rewetting the subsample with distilled water when it is time for analysis. The second method allows for easier storage.

For the current study, the significance of the method by which the subsample was presented to the Sympatec LIXELL image analysis instrument was only realized in the last year of sampling (2014). During the final year of sampling, suspended sediment samples were kept in their original solutions and compared to subsamples prepared during earlier years that had been dried and rewet in order to determine if there was a significant difference between the two methods.

Table A.1. P-values for particle size for subsamples: 1) kept wet and 2) dried and rewet with distilled water

Wet v.s. Dried and Rewet Sample P-values for Particle Size						
/olume and Area Classe (Percentile)	Volume (Percentile)	Area (Percentile)	Mean Volume Surface Area	Mean Mass Surface Area	Size Fraction Classes (Percentile)	Size Fraction (Percentile)
10	<0.001	<0.001	2.15E-10	8.62E-10	2-62um	0.98
25	<0.001	<0.001			62-600um	0.835
50	<0.001	<0.001			>600pm	0.004
75	<0.001	<0.001				
90	<0.001	<0.001				

Table A.2. P-values for particle shape for subsamples: 1) kept wet and 2) dried and rewet with distilled water

Shape P-value for samples kept wet v.s. dried and rewet		
50% Aspect	50% Sphericity	50% Convexity
<0.0001	<0.0001	<0.0001

Results suggest that there is a significant difference among most size and shape variables between the two methods used for suspended sediment preparation. This presents serious complications for the present study as well as all other studies that utilize particle size and shape fingerprints for source discrimination. Although it is ideal to keep suspended sediment subsamples in solution from the time they are collected to when they are analyzed, preferably with water from the channel in which they were collected, this was not possible because samples collected from 2008-2013 were dried and rewet with distilled water, with only those collected in 2014 remaining in solution. Therefore it was decided that all samples would be dried and rewet to ensure consistency among results.

Appendix B: MixSIAR spreadsheet design for Single Watershed Source Sampling

Table A.3. MixSIAR mixture spreadsheet for Single Watershed Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137 divided by categorical covariate "Year"

Year	V	R	L	U	B	G	Cs-137	X	Z
1	9.2	99.6	62.7	20.3	53.4	78.2	10.5	37.3	6.4
1	8.5	96.3	62.5	18.0	55.3	77.8	9.9	36.6	6.7
1	9.2	100.6	63.0	20.2	54.3	79.1	16.3	37.7	6.5
1	8.8	97.5	62.8	18.4	55.4	78.9	8.7	37.2	6.7
1	9.1	100.1	63.1	19.8	54.5	79.4	6.8	37.7	6.5
1	8.3	87.7	59.3	18.9	47.7	68.5	5.8	32.7	5.7
1	8.0	84.5	58.8	17.6	47.6	67.2	6.6	31.8	5.8
1	7.8	82.8	58.4	17.0	47.2	66.0	9.3	31.2	5.7
1	7.9	83.8	58.4	17.8	46.8	66.0	8.9	31.3	5.7
1	7.7	80.8	57.5	17.5	45.2	63.6	6.1	30.2	5.5
1	7.8	82.8	58.1	17.5	46.5	65.2	2.2	30.9	5.6
1	7.5	78.3	56.3	17.6	43.0	60.4	13.6	28.9	5.2
1	7.6	78.9	56.5	17.7	43.3	61.0	9.1	29.1	5.2
1	8.2	87.7	59.3	18.5	48.2	68.4	22.7	32.6	5.8
2	7.4	77.4	56.2	17.1	43.2	60.1	14.3	28.6	5.3
2	7.4	76.7	55.8	17.4	42.2	59.1	11.4	28.2	5.1
2	8.1	89.4	60.0	18.1	50.1	70.4	8.9	33.4	6.1
2	7.3	74.8	55.2	17.1	41.4	57.7	8.1	27.6	5.0
2	7.4	78.0	56.3	17.4	43.3	60.4	7.2	28.8	5.3
2	7.7	79.9	56.9	17.9	43.8	61.8	5.4	29.5	5.3
2	7.4	77.4	56.2	17.2	43.1	60.1	10.3	28.7	5.2
2	7.4	77.5	56.1	17.2	43.1	60.0	5.3	28.6	5.2
3	8.6	91.0	60.3	19.2	49.5	71.3	4.3	34.0	5.9
3	8.0	86.0	59.2	17.6	48.6	68.2	15.2	32.3	5.9
3	8.0	84.0	58.5	17.9	46.9	66.3	10.3	31.5	5.7
3	9.0	97.9	62.7	19.2	54.1	78.3	11.3	37.1	6.5
3	9.0	99.2	63.2	18.9	55.8	80.1	4.5	37.8	6.7
3	8.6	88.8	59.3	20.0	46.7	68.3	4.1	32.8	5.6
3	7.7	80.9	57.5	17.5	45.2	63.5	3.8	30.2	5.5
3	7.6	77.4	56.1	17.6	42.5	60.0	9.7	28.6	5.1
3	7.9	82.1	57.5	18.3	44.7	63.6	13.0	30.4	5.4
4	9.2	99.4	62.5	20.6	52.6	77.5	9.8	37.0	6.3
4	8.5	92.8	61.3	18.2	52.2	74.3	11.0	35.1	6.3
4	8.1	83.3	57.8	18.6	45.0	64.4	8.6	30.8	5.4
4	8.6	92.4	61.1	18.6	51.4	73.8	3.9	34.9	6.2
4	8.9	91.7	60.6	19.7	49.1	72.0	5.1	34.3	5.9
4	8.9	98.2	62.5	19.5	53.9	77.7	5.0	36.9	6.5
4	7.7	76.5	55.8	18.1	41.2	59.1	10.0	28.3	5.0
4	7.6	77.6	56.2	17.6	42.7	60.3	7.3	28.8	5.2
4	7.7	79.8	56.9	18.0	43.7	61.9	6.0	29.6	5.3
5	6.9	71.0	53.9	16.6	39.4	54.5	10.7	26.0	4.8
5	6.8	70.2	53.6	16.5	38.9	53.7	4.9	25.7	4.8
5	8.8	96.0	62.2	19.1	53.3	76.8	8.7	36.4	6.4
5	8.6	95.2	62.2	18.3	54.4	77.0	7.4	36.3	6.6
5	9.1	101.1	64.0	18.9	57.7	82.5	9.6	38.9	6.9
5	9.4	103.6	64.0	20.4	56.2	82.3	7.5	39.1	6.7
5	9.2	101.2	63.4	19.9	55.3	80.5	13.7	38.2	6.6
5	9.5	100.4	62.7	21.2	52.4	78.1	0.1	37.4	6.2
6	8.9	92.5	60.8	19.8	49.5	72.5	13.3	34.6	5.9
6	8.9	92.2	60.7	19.7	49.5	72.4	6.8	34.5	5.9
6	9.0	98.3	62.6	19.5	53.9	78.1	5.4	37.0	6.5
6	7.8	80.0	57.2	17.5	44.6	63.0	7.1	29.9	5.4
6	7.5	76.3	56.0	17.2	42.5	59.9	3.5	28.5	5.2
6	8.4	89.1	59.7	19.1	48.4	69.5	5.1	33.2	5.8
7	6.5	64.8	51.6	16.0	35.8	49.1	8.1	23.6	4.4

Table A.4. MixSIAR source spreadsheet for Single Watershed Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137

Source	MeanV	SDV	MeanR	SDR	MeanL	SDL	MeanU	SDU	MeanB	SDB	MeanG	SDG	MeanCs-137	SDCs-137	MeanX	SDX	MeanZ	SDZ	n
Cultivated Topsoil	8.709047	1.485543	89.94995	15.70947	59.02725	5.426791	20.57639	2.777642	46.02929	8.355251	69.63631	12.40202	6.23810724	3.763303	33.50427	5.819212	5.576122	0.923142	44
Forest Topsoil	6.785061	1.564927	82.68756	27.39083	56.19025	10.46288	17.52571	2.802449	46.66171	19.15614	63.36084	24.45143	14.5245332	8.538175	30.30946	11.20455	5.770278	2.426395	13
Cultivated Streambank	7.616282	0.649594	76.77787	6.581039	55.45277	2.178998	18.32032	1.817734	40.77938	3.407218	58.30965	5.036837	7.16743657	9.611818	27.90249	2.381883	4.976653	0.351034	71
Forest Streambank	5.13456	0.294642	44.13392	3.295329	41.0457	1.124555	16.37055	1.194383	20.62663	0.921739	29.00747	1.668754	17.8392046	9.918352	14.53432	0.925015	2.527694	0.103836	8

Table A.5. MixSIAR source spreadsheet for Single Watershed Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137

Source	MeanV	SDV	MeanR	SDR	MeanL	SDL	MeanU	SDU	MeanB	SDB	MeanG	SDG	MeanCs-137	SDCs-137	MeanX	SDX	MeanZ	SDZ	n
Cultivated Topsoil	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	44
Forest Topsoil	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	13
Cultivated Streambank	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	71
Forest Streambank	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	8

Appendix C: MixSIAR spreadsheet design for Regional Source Sampling

Table A.6. MixSIAR mixture spreadsheet for Regional Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137 divided by categorical covariate "Year"

Year	V	R	L	U	B	G	Cs-137	X	Z
1	9.2	99.6	62.7	20.3	53.4	78.2	10.5	37.3	6.4
1	8.5	96.3	62.5	18.0	55.3	77.8	9.9	36.6	6.7
1	9.2	100.6	63.0	20.2	54.3	79.1	16.3	37.7	6.5
1	8.8	97.5	62.8	18.4	55.4	78.9	8.7	37.2	6.7
1	9.1	100.1	63.1	19.8	54.5	79.4	6.8	37.7	6.5
1	8.3	87.7	59.3	18.9	47.7	68.5	5.8	32.7	5.7
1	8.0	84.5	58.8	17.6	47.6	67.2	6.6	31.8	5.8
1	7.8	82.8	58.4	17.0	47.2	66.0	9.3	31.2	5.7
1	7.9	83.8	58.4	17.8	46.8	66.0	8.9	31.3	5.7
1	7.7	80.8	57.5	17.5	45.2	63.6	6.1	30.2	5.5
1	7.8	82.8	58.1	17.5	46.5	65.2	2.2	30.9	5.6
1	7.5	78.3	56.3	17.6	43.0	60.4	13.6	28.9	5.2
1	7.6	78.9	56.5	17.7	43.3	61.0	9.1	29.1	5.2
1	8.2	87.7	59.3	18.5	48.2	68.4	22.7	32.6	5.8
2	7.4	77.4	56.2	17.1	43.2	60.1	14.3	28.6	5.3
2	7.4	76.7	55.8	17.4	42.2	59.1	11.4	28.2	5.1
2	8.1	89.4	60.0	18.1	50.1	70.4	8.9	33.4	6.1
2	7.3	74.8	55.2	17.1	41.4	57.7	8.1	27.6	5.0
2	7.4	78.0	56.3	17.4	43.3	60.4	7.2	28.8	5.3
2	7.7	79.9	56.9	17.9	43.8	61.8	5.4	29.5	5.3
2	7.4	77.4	56.2	17.2	43.1	60.1	10.3	28.7	5.2
2	7.4	77.5	56.1	17.2	43.1	60.0	5.3	28.6	5.2
3	8.6	91.0	60.3	19.2	49.5	71.3	4.3	34.0	5.9
3	8.0	86.0	59.2	17.6	48.6	68.2	15.2	32.3	5.9
3	8.0	84.0	58.5	17.9	46.9	66.3	10.3	31.5	5.7
3	9.0	97.9	62.7	19.2	54.1	78.3	11.3	37.1	6.5
3	9.0	99.2	63.2	18.9	55.8	80.1	4.5	37.8	6.7
3	8.6	88.8	59.3	20.0	46.7	68.3	4.1	32.8	5.6
3	7.7	80.9	57.5	17.5	45.2	63.5	3.8	30.2	5.5
3	7.6	77.4	56.1	17.6	42.5	60.0	9.7	28.6	5.1
3	7.9	82.1	57.5	18.3	44.7	63.6	13.0	30.4	5.4
4	9.2	99.4	62.5	20.6	52.6	77.5	9.8	37.0	6.3
4	8.5	92.8	61.3	18.2	52.2	74.3	11.0	35.1	6.3
4	8.1	83.3	57.8	18.6	45.0	64.4	8.6	30.8	5.4
4	8.6	92.4	61.1	18.6	51.4	73.8	3.9	34.9	6.2
4	8.9	91.7	60.6	19.7	49.1	72.0	5.1	34.3	5.9
4	8.9	98.2	62.5	19.5	53.9	77.7	5.0	36.9	6.5
4	7.7	76.5	55.8	18.1	41.2	59.1	10.0	28.3	5.0
4	7.6	77.6	56.2	17.6	42.7	60.3	7.3	28.8	5.2
4	7.7	79.8	56.9	18.0	43.7	61.9	6.0	29.6	5.3
5	6.9	71.0	53.9	16.6	39.4	54.5	10.7	26.0	4.8
5	6.8	70.2	53.6	16.5	38.9	53.7	4.9	25.7	4.8
5	8.8	96.0	62.2	19.1	53.3	76.8	8.7	36.4	6.4
5	8.6	95.2	62.2	18.3	54.4	77.0	7.4	36.3	6.6
5	9.1	101.1	64.0	18.9	57.7	82.5	9.6	38.9	6.9
5	9.4	103.6	64.0	20.4	56.2	82.3	7.5	39.1	6.7
5	9.2	101.2	63.4	19.9	55.3	80.5	13.7	38.2	6.6
5	9.5	100.4	62.7	21.2	52.4	78.1	0.1	37.4	6.2
6	8.9	92.5	60.8	19.8	49.5	72.5	13.3	34.6	5.9
6	8.9	92.2	60.7	19.7	49.5	72.4	6.8	34.5	5.9
6	9.0	98.3	62.6	19.5	53.9	78.1	5.4	37.0	6.5
6	7.8	80.0	57.2	17.5	44.6	63.0	7.1	29.9	5.4
6	7.5	76.3	56.0	17.2	42.5	59.9	3.5	28.5	5.2
6	8.4	89.1	59.7	19.1	48.4	69.5	5.1	33.2	5.8
7	6.5	64.8	51.6	16.0	35.8	49.1	8.1	23.6	4.4

Table A.7. MixSIAR source spreadsheet for Regional Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137

	MeanV	SDV	MeanR	SDR	MeanL	SDL	MeanU	SDU	MeanB	SDB	MeanG	SDG	MeanCs-137	SDCs-137	MeanX	SDX	MeanZ	SDZ	n
Cultivated Topsoil	8.7	1.5	89.9	15.7	59.0	5.4	20.6	2.8	46.0	8.4	68.5	13.4	6.2	3.8	33.0	6.3	5.5	1.0	44
Forest Topsoil	6.5	1.5	77.1	27.3	54.3	10.2	17.0	2.8	43.3	18.6	58.7	24.1	19.3	6.2	28.1	11.1	5.4	2.3	16
Cultivated Streambank	7.6	0.6	76.8	6.6	55.5	2.2	18.3	1.8	40.8	3.4	58.3	5.0	7.2	9.6	27.9	2.4	5.0	0.4	71
Forest Streambank	5.2	0.6	52.5	8.1	45.7	4.3	15.0	2.0	28.2	6.8	37.7	8.0	13.3	8.0	18.3	3.5	3.5	0.9	18

Table A.8. MixSIAR discriminant spreadsheet for Regional Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137

	MeanV	SDV	MeanR	SDR	MeanL	SDL	MeanU	SDU	MeanB	SDB	MeanG	SDG	MeanCs-137	SDCs-137	MeanX	SDX	MeanZ	SDZ	n
Cultivated Topsoil	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	44
Forest Topsoil	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	16
Cultivated Streambank	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	71
Forest Streambank	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	18

Appendix D: MixSIAR spreadsheet design for Multiple Watershed Source Sampling

Table A.9. MixSIAR mixture spreadsheet for Multiple Watershed Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137 divided by categorical covariates "Year" and "Site"

Site	Year	V	R	L	U	B	G	Cs-137	X	Z
W1	1	9.2	99.6	62.7	20.3	53.4	78.2	10.5	37.3	6.4
W1	1	8.5	96.3	62.5	18.0	55.3	77.8	9.9	36.6	6.7
W1	1	8.8	97.5	62.8	18.4	55.4	78.9	8.7	37.2	6.7
W1	1	8.3	87.7	59.3	18.9	47.7	68.5	5.8	32.7	5.7
W1	1	8.0	84.5	58.8	17.6	47.6	67.2	6.6	31.8	5.8
W1	1	7.9	83.8	58.4	17.8	46.8	66.0	8.9	31.3	5.7
W1	1	7.7	80.8	57.5	17.5	45.2	63.6	6.1	30.2	5.5
W1	1	7.5	78.3	56.3	17.6	43.0	60.4	13.6	28.9	5.2
W1	1	7.6	78.9	56.5	17.7	43.3	61.0	9.1	29.1	5.2
W1	2	7.4	77.4	56.2	17.1	43.2	60.1	14.3	28.6	5.3
W1	2	7.4	76.7	55.8	17.4	42.2	59.1	11.4	28.2	5.1
W1	2	7.3	74.8	55.2	17.1	41.4	57.7	8.1	27.6	5.0
W1	2	7.4	78.0	56.3	17.4	43.3	60.4	7.2	28.8	5.3
W1	2	7.7	79.9	56.9	17.9	43.8	61.8	5.4	29.5	5.3
W1	2	7.4	77.4	56.2	17.2	43.1	60.1	10.3	28.7	5.2
W1	3	8.6	91.0	60.3	19.2	49.5	71.3	4.3	34.0	5.9
W1	3	8.0	86.0	59.2	17.6	48.6	68.2	15.2	32.3	5.9
W1	3	9.0	97.9	62.7	19.2	54.1	78.3	11.3	37.1	6.5
W1	3	9.0	99.2	63.2	18.9	55.8	80.1	4.5	37.8	6.7
W1	3	7.7	80.9	57.5	17.5	45.2	63.5	3.8	30.2	5.5
W1	3	7.6	77.4	56.1	17.6	42.5	60.0	9.7	28.6	5.1
W1	4	9.2	99.4	62.5	20.6	52.6	77.5	9.8	37.0	6.3
W1	4	8.5	92.8	61.3	18.2	52.2	74.3	11.0	35.1	6.3
W1	4	8.6	92.4	61.1	18.6	51.4	73.8	3.9	34.9	6.2
W1	4	8.9	91.7	60.6	19.7	49.1	72.0	5.1	34.3	5.9
W1	4	7.7	76.5	55.8	18.1	41.2	59.1	10.0	28.3	5.0
W1	4	7.6	77.6	56.2	17.6	42.7	60.3	7.3	28.8	5.2
W1	5	6.9	71.0	53.9	16.6	39.4	54.5	10.7	26.0	4.8
W1	5	6.8	70.2	53.6	16.5	38.9	53.7	4.9	25.7	4.8
W1	5	8.8	96.0	62.2	19.1	53.3	76.8	8.7	36.4	6.4
W1	5	8.6	95.2	62.2	18.3	54.4	77.0	7.4	36.3	6.6
W1	5	9.4	103.6	64.0	20.4	56.2	82.3	7.5	39.1	6.7
W1	5	9.2	101.2	63.4	19.9	55.3	80.5	13.7	38.2	6.6
W1	6	8.9	92.5	60.8	19.8	49.5	72.5	13.3	34.6	5.9
W1	6	8.9	92.2	60.7	19.7	49.5	72.4	6.8	34.5	5.9
W1	6	7.8	80.0	57.2	17.5	44.6	63.0	7.1	29.9	5.4
W1	6	7.5	76.3	56.0	17.2	42.5	59.9	3.5	28.5	5.2
W1	7	6.5	64.8	51.6	16.0	35.8	49.1	8.1	23.6	4.4
W1	7	6.7	69.1	53.1	16.4	38.2	52.5	5.8	25.1	4.7
W1	7	8.9	95.3	62.1	18.9	53.1	76.6	7.0	36.2	6.4
W1	7	8.9	96.6	62.2	19.4	53.1	76.7	1.8	36.4	6.4
W1	7	9.2	95.7	61.6	20.5	50.7	74.9	11.5	35.8	6.0
W1	7	9.0	92.5	60.8	19.8	49.6	72.7	3.6	34.7	5.9
W1	1	9.2	100.6	63.0	20.2	54.3	79.1	16.3	37.7	6.5
W1	1	9.1	100.1	63.1	19.8	54.5	79.4	6.8	37.7	6.5
W1	1	7.8	82.8	58.4	17.0	47.2	66.0	9.3	31.2	5.7
W1	1	7.8	82.8	58.1	17.5	46.5	65.2	2.2	30.9	5.6
W1	1	8.2	87.7	59.3	18.5	48.2	68.4	22.7	32.6	5.8
W1	2	8.1	89.4	60.0	18.1	50.1	70.4	8.9	33.4	6.1
W1	2	7.4	77.5	56.1	17.2	43.1	60.0	5.3	28.6	5.2
W1	3	8.0	84.0	58.5	17.9	46.9	66.3	10.3	31.5	5.7
W1	3	8.6	88.8	59.3	20.0	46.7	68.3	4.1	32.8	5.6
W1	3	7.9	82.1	57.5	18.3	44.7	63.6	13.0	30.4	5.4
W1	4	8.1	83.3	57.8	18.6	45.0	64.4	8.6	30.8	5.4
W1	4	8.9	98.2	62.5	19.5	53.9	77.7	5.0	36.9	6.5
W1	4	7.7	79.8	56.9	18.0	43.7	61.9	6.0	29.6	5.3
W1	5	9.1	101.1	64.0	18.9	57.7	82.5	9.6	38.9	6.9
W1	5	9.5	100.4	62.7	21.2	52.4	78.1	0.1	37.4	6.2
W1	6	9.0	98.3	62.6	19.5	53.9	78.1	5.4	37.0	6.5
W1	6	8.4	89.1	59.7	19.1	48.4	69.5	5.1	33.2	5.8
W1	7	9.5	98.9	62.4	21.2	51.6	77.4	1.1	37.0	6.1
W1	7	9.2	98.8	62.5	20.4	52.9	77.7	0.0	37.1	6.3
W2	1	8.2	87.8	59.3	18.7	47.9	68.4	15.9	32.6	5.8
W2	1	7.3	83.1	58.5	16.0	49.1	66.4	1.5	31.3	6.0
W2	1	6.7	72.3	54.4	16.1	41.0	55.6	2.7	26.5	5.0
W2	3	9.0	92.7	60.5	20.5	48.3	71.6	8.2	34.3	5.8
W2	3	7.4	74.6	54.9	17.7	40.2	56.8	9.0	27.2	4.9
W2	4	8.3	84.9	58.2	19.2	45.0	65.2	9.9	31.2	5.4
W2	4	7.3	72.3	54.1	17.6	38.7	54.9	13.0	26.4	4.7
W2	4	7.5	75.4	55.4	17.8	40.9	58.0	15.7	27.8	5.0
W2	5	5.9	69.3	53.6	14.7	41.0	53.8	7.5	25.5	5.1
W2	5	8.7	97.9	64.9	13.4	64.1	86.4	0.0	39.2	7.7
W2	5	8.5	89.4	59.7	19.3	48.1	69.6	5.7	33.2	5.8
W2	6	8.5	91.3	60.5	18.9	50.0	71.9	5.9	34.1	6.0
W2	6	7.2	72.8	54.5	17.1	40.0	56.1	12.8	26.8	4.9
W2	7	6.1	61.3	50.0	15.8	33.5	45.7	6.3	22.0	4.1
W2	7	9.0	94.8	61.3	20.0	50.7	74.1	6.9	35.4	6.1
W2	7	9.1	96.6	62.1	20.2	52.0	76.3	7.3	36.4	6.2
W3	1	7.9	85.2	58.5	18.1	46.9	66.3	6.3	31.6	5.7
W3	1	7.5	79.0	56.6	17.6	43.5	61.1	5.1	29.2	5.3
W3	2	5.8	56.9	47.9	16.0	30.1	41.2	22.5	20.0	3.7
W3	2	5.8	55.0	47.2	15.7	29.2	40.0	16.1	19.4	3.6
W3	3	8.9	90.2	59.7	20.4	46.6	69.3	8.0	33.2	5.6
W3	3	8.4	82.0	56.9	20.1	41.6	61.6	13.2	29.8	5.0
W3	4	8.9	92.7	60.7	19.9	49.2	72.3	10.9	34.5	5.9
W3	4	9.4	94.1	60.5	21.9	47.0	71.6	10.1	34.5	5.6
W3	6	8.7	96.4	62.0	19.2	52.9	76.1	7.3	36.1	6.4
W3	6	6.9	70.5	53.5	16.9	38.6	53.5	10.3	25.7	4.7
W3	7	5.7	54.6	47.3	15.3	29.5	40.1	8.3	19.4	3.6
W3	7	8.6	87.8	59.1	19.5	46.2	67.8	11.1	32.4	5.5
W3	7	8.2	84.9	58.3	18.9	45.5	65.8	0.3	31.4	5.5
W4	1	6.0	56.7	47.2	17.1	28.5	39.8	29.1	19.5	3.5
W4	1	5.4	49.6	43.7	16.9	23.8	33.2	22.7	16.6	2.9

Table A.10. MixSIAR source spreadsheet for Multiple Watershed Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137

Source	Site	MeanV	SDV	MeanR	SDR	MeanL	SDL	MeanU	SDU	MeanB	SDB	MeanG	SDG	MeanCs-137	SDCs-137	MeanX	SDX	MeanZ	SDZ	n
Cultivated Topsoil	W1	8.7	1.5	89.9	15.7	59.0	5.4	20.6	2.8	46.0	8.4	69.6	12.4	6.2	3.8	33.5	5.8	5.6	0.9	44
Cultivated Topsoil	W2	8.7	1.5	89.9	15.7	59.0	5.4	20.6	2.8	46.0	8.4	69.6	12.4	6.2	3.8	33.5	5.8	5.6	0.9	44
Cultivated Topsoil	W3	8.7	1.5	89.9	15.7	59.0	5.4	20.6	2.8	46.0	8.4	69.6	12.4	6.2	3.8	33.5	5.8	5.6	0.9	44
Cultivated Topsoil	W4	8.7	1.5	89.9	15.7	59.0	5.4	20.6	2.8	46.0	8.4	69.6	12.4	6.2	3.8	33.5	5.8	5.6	0.9	44
Cultivated Topsoil	W8	8.7	1.5	89.9	15.7	59.0	5.4	20.6	2.8	46.0	8.4	69.6	12.4	6.2	3.8	33.5	5.8	5.6	0.9	44
Forest Topsoil	W1	6.8	1.6	82.7	27.4	56.2	10.5	17.5	2.8	46.7	19.2	63.4	24.5	14.5	8.5	30.3	11.2	5.8	2.4	13
Forest Topsoil	W2	6.8	1.6	82.7	27.4	56.2	10.5	17.5	2.8	46.7	19.2	63.4	24.5	14.5	8.5	30.3	11.2	5.8	2.4	13
Forest Topsoil	W3	6.8	1.6	82.7	27.4	56.2	10.5	17.5	2.8	46.7	19.2	63.4	24.5	14.5	8.5	30.3	11.2	5.8	2.4	13
Forest Topsoil	W4	6.8	1.6	82.7	27.4	56.2	10.5	17.5	2.8	46.7	19.2	63.4	24.5	14.5	8.5	30.3	11.2	5.8	2.4	13
Forest Topsoil	W8	6.8	1.6	82.7	27.4	56.2	10.5	17.5	2.8	46.7	19.2	63.4	24.5	14.5	8.5	30.3	11.2	5.8	2.4	13
Cultivated Streambank	W1	7.6	0.6	76.8	6.6	55.5	2.2	18.3	1.8	40.8	3.4	58.3	5.0	7.2	9.6	27.9	2.4	5.0	0.4	71
Cultivated Streambank	W2	7.6	0.6	76.8	6.6	55.5	2.2	18.3	1.8	40.8	3.4	58.3	5.0	7.2	9.6	27.9	2.4	5.0	0.4	71
Cultivated Streambank	W3	7.6	0.6	76.8	6.6	55.5	2.2	18.3	1.8	40.8	3.4	58.3	5.0	7.2	9.6	27.9	2.4	5.0	0.4	71
Cultivated Streambank	W4	7.6	0.6	76.8	6.6	55.5	2.2	18.3	1.8	40.8	3.4	58.3	5.0	7.2	9.6	27.9	2.4	5.0	0.4	71
Cultivated Streambank	W8	7.6	0.6	76.8	6.6	55.5	2.2	18.3	1.8	40.8	3.4	58.3	5.0	7.2	9.6	27.9	2.4	5.0	0.4	71
Forest Streambank	W1	5.1	0.3	44.1	3.3	41.0	1.1	16.4	1.2	20.6	0.9	29.0	1.7	17.8	9.9	14.5	0.9	2.5	0.1	8
Forest Streambank	W2	5.1	0.3	44.1	3.3	41.0	1.1	16.4	1.2	20.6	0.9	29.0	1.7	17.8	9.9	14.5	0.9	2.5	0.1	8
Forest Streambank	W3	5.1	0.3	44.1	3.3	41.0	1.1	16.4	1.2	20.6	0.9	29.0	1.7	17.8	9.9	14.5	0.9	2.5	0.1	8
Forest Streambank	W4	5.1	0.3	44.1	3.3	41.0	1.1	16.4	1.2	20.6	0.9	29.0	1.7	17.8	9.9	14.5	0.9	2.5	0.1	8
Forest Streambank	W8	5.1	0.3	44.1	3.3	41.0	1.1	16.4	1.2	20.6	0.9	29.0	1.7	17.8	9.9	14.5	0.9	2.5	0.1	8

Table A.11. MixSIAR source spreadsheet for Multiple Watershed Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137

Source	MeanV	SDV	MeanR	SDR	MeanL	SDL	MeanU	SDU	MeanB	SDB	MeanG	SDG	MeanCs-137	SDCs-137	MeanX	SDX	MeanZ	SDZ
Cultivated Topsoil	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Forst Topsoil	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Cultivated Streambank	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Forest Streambank	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0

Appendix E: MixSIAR spreadsheet design for Upstream Source Sampling

Table A.12. MixSIAR mixture spreadsheet for Upstream Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137 divided by categorical covariates “Year” and “Site”

Site	Year	V	R	L	U	B	G	Cs-137	X	Z
W1	1	9.2	99.6	62.7	20.3	53.4	78.2	10.5	37.3	6.4
W1	1	8.5	96.3	62.5	18.0	55.3	77.8	9.9	36.6	6.7
W1	1	8.8	97.5	62.8	18.4	55.4	78.9	8.7	37.2	6.7
W1	1	8.3	87.7	59.3	18.9	47.7	68.5	5.8	32.7	5.7
W1	1	8.0	84.5	58.8	17.6	47.6	67.2	6.6	31.8	5.8
W1	1	7.9	83.8	58.4	17.8	46.8	66.0	8.9	31.3	5.7
W1	1	7.7	80.8	57.5	17.5	45.2	63.6	6.1	30.2	5.5
W1	1	7.5	78.3	56.3	17.6	43.0	60.4	13.6	28.9	5.2
W1	1	7.6	78.9	56.5	17.7	43.3	61.0	9.1	29.1	5.2
W1	2	7.4	77.4	56.2	17.1	43.2	60.1	14.3	28.6	5.3
W1	2	7.4	76.7	55.8	17.4	42.2	59.1	11.4	28.2	5.1
W1	2	7.3	74.8	55.2	17.1	41.4	57.7	8.1	27.6	5.0
W1	2	7.4	78.0	56.3	17.4	43.3	60.4	7.2	28.8	5.3
W1	2	7.7	79.9	56.9	17.9	43.8	61.8	5.4	29.5	5.3
W1	2	7.4	77.4	56.2	17.2	43.1	60.1	10.3	28.7	5.2
W1	3	8.6	91.0	60.3	19.2	49.5	71.3	4.3	34.0	5.9
W1	3	8.0	86.0	59.2	17.6	48.6	68.2	15.2	32.3	5.9
W1	3	9.0	97.9	62.7	19.2	54.1	78.3	11.3	37.1	6.5
W1	3	9.0	99.2	63.2	18.9	55.8	80.1	4.5	37.8	6.7
W1	3	7.7	80.9	57.5	17.5	45.2	63.5	3.8	30.2	5.5
W1	3	7.6	77.4	56.1	17.6	42.5	60.0	9.7	28.6	5.1
W1	4	9.2	99.4	62.5	20.6	52.6	77.5	9.8	37.0	6.3
W1	4	8.5	92.8	61.3	18.2	52.2	74.3	11.0	35.1	6.3
W1	4	8.6	92.4	61.1	18.6	51.4	73.8	3.9	34.9	6.2
W1	4	8.9	91.7	60.6	19.7	49.1	72.0	5.1	34.3	5.9
W1	4	7.7	76.5	55.8	18.1	41.2	59.1	10.0	28.3	5.0
W1	4	7.6	77.6	56.2	17.6	42.7	60.3	7.3	28.8	5.2
W1	5	6.9	71.0	53.9	16.6	39.4	54.5	10.7	26.0	4.8
W1	5	6.8	70.2	53.6	16.5	38.9	53.7	4.9	25.7	4.8
W1	5	8.8	96.0	62.2	19.1	53.3	76.8	8.7	36.4	6.4
W1	5	8.6	95.2	62.2	18.3	54.4	77.0	7.4	36.3	6.6
W1	5	9.4	103.6	64.0	20.4	56.2	82.3	7.5	39.1	6.7
W1	5	9.2	101.2	63.4	19.9	55.3	80.5	13.7	38.2	6.6
W1	6	8.9	92.5	60.8	19.8	49.5	72.5	13.3	34.6	5.9
W1	6	8.9	92.2	60.7	19.7	49.5	72.4	6.8	34.5	5.9
W1	6	7.8	80.0	57.2	17.5	44.6	63.0	7.1	29.9	5.4
W1	6	7.5	76.3	56.0	17.2	42.5	59.9	3.5	28.5	5.2
W1	7	6.5	64.8	51.6	16.0	35.8	49.1	8.1	23.6	4.4
W1	7	6.7	69.1	53.1	16.4	38.2	52.5	5.8	25.1	4.7
W1	7	8.9	95.3	62.1	18.9	53.1	76.6	7.0	36.2	6.4
W1	7	8.9	96.6	62.2	19.4	53.1	76.7	1.8	36.4	6.4
W1	7	9.2	95.7	61.6	20.5	50.7	74.9	11.5	35.8	6.0
W1	7	9.0	92.5	60.8	19.8	49.6	72.7	3.6	34.7	5.9
W1	1	9.2	100.6	63.0	20.2	54.3	79.1	16.3	37.7	6.5
W1	1	9.1	100.1	63.1	19.8	54.5	79.4	6.8	37.7	6.5
W1	1	7.8	82.8	58.4	17.0	47.2	66.0	9.3	31.2	5.7
W1	1	7.8	82.8	58.1	17.5	46.5	65.2	2.2	30.9	5.6
W1	1	8.2	87.7	59.3	18.5	48.2	68.4	22.7	32.6	5.8
W1	2	8.1	89.4	60.0	18.1	50.1	70.4	8.9	33.4	6.1
W1	2	7.4	77.5	56.1	17.2	43.1	60.0	5.3	28.6	5.2
W1	3	8.0	84.0	58.5	17.9	46.9	66.3	10.3	31.5	5.7
W1	3	8.6	88.8	59.3	20.0	46.7	68.3	4.1	32.8	5.6
W1	3	7.9	82.1	57.5	18.3	44.7	63.6	13.0	30.4	5.4
W1	4	8.1	83.3	57.8	18.6	45.0	64.4	8.6	30.8	5.4
W1	4	8.9	98.2	62.5	19.5	53.9	77.7	5.0	36.9	6.5
W1	4	7.7	79.8	56.9	18.0	43.7	61.9	6.0	29.6	5.3
W1	5	9.1	101.1	64.0	18.9	57.7	82.5	9.6	38.9	6.9
W1	5	9.5	100.4	62.7	21.2	52.4	78.1	0.1	37.4	6.2
W1	6	9.0	98.3	62.6	19.5	53.9	78.1	5.4	37.0	6.5
W1	6	8.4	89.1	59.7	19.1	48.4	69.5	5.1	33.2	5.8
W1	7	9.5	98.9	62.4	21.2	51.6	77.4	1.1	37.0	6.1
W1	7	9.2	98.8	62.5	20.4	52.9	77.7	0.0	37.1	6.3
W2	1	8.2	87.8	59.3	18.7	47.9	68.4	15.9	32.6	5.8
W2	1	7.3	83.1	58.5	16.0	49.1	66.4	1.5	31.3	6.0
W2	1	6.7	72.3	54.4	16.1	41.0	55.6	2.7	26.5	5.0
W2	3	9.0	92.7	60.5	20.5	48.3	71.6	8.2	34.3	5.8
W2	3	7.4	74.6	54.9	17.7	40.2	56.8	9.0	27.2	4.9
W2	4	8.3	84.9	58.2	19.2	45.0	65.2	9.9	31.2	5.4
W2	4	7.3	72.3	54.1	17.6	38.7	54.9	13.0	26.4	4.7
W2	4	7.5	75.4	55.4	17.8	40.9	58.0	15.7	27.8	5.0
W2	5	5.9	69.3	53.6	14.7	41.0	53.8	7.5	25.5	5.1
W2	5	8.7	97.9	64.9	13.4	64.1	86.4	0.0	39.2	7.7
W2	5	8.5	89.4	59.7	19.3	48.1	69.6	5.7	33.2	5.8
W2	6	8.5	91.3	60.5	18.9	50.0	71.9	5.9	34.1	6.0
W2	6	7.2	72.8	54.5	17.1	40.0	56.1	12.8	26.8	4.9
W2	7	6.1	61.3	50.0	15.8	33.5	45.7	6.3	22.0	4.1
W2	7	9.0	94.8	61.3	20.0	50.7	74.1	6.9	35.4	6.1
W2	7	9.1	96.6	62.1	20.2	52.0	76.3	7.3	36.4	6.2
W3	1	7.9	85.2	58.5	18.1	46.9	66.3	6.3	31.6	5.7
W3	1	7.5	79.0	56.6	17.6	43.5	61.1	5.1	29.2	5.3
W3	2	5.8	56.9	47.9	16.0	30.1	41.2	22.5	20.0	3.7
W3	2	5.8	55.0	47.2	15.7	29.2	40.0	16.1	19.4	3.6
W3	3	8.9	90.2	59.7	20.4	46.6	69.3	8.0	33.2	5.6
W3	3	8.4	82.0	56.9	20.1	41.6	61.6	13.2	29.8	5.0
W3	4	8.9	92.7	60.7	19.9	49.2	72.3	10.9	34.5	5.9
W3	4	9.4	94.1	60.5	21.9	47.0	71.6	10.1	34.5	5.6
W3	6	8.7	96.4	62.0	19.2	52.9	76.1	7.3	36.1	6.4
W3	6	6.9	70.5	53.5	16.9	38.6	53.5	10.3	25.7	4.7
W3	7	5.7	54.6	47.3	15.3	29.5	40.1	8.3	19.4	3.6
W3	7	8.6	87.8	59.1	19.5	46.2	67.8	11.1	32.4	5.5
W3	7	8.2	84.9	58.3	18.9	45.5	65.8	0.3	31.4	5.5
W4	1	6.0	56.7	47.2	17.1	28.5	39.8	29.1	19.5	3.5
W4	1	5.4	49.6	43.7	16.9	23.8	33.2	22.7	16.6	2.9

Table A.13. MixSIAR source spreadsheet for Upstream Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137

Source	Site	MeanV	SDV	MeanR	SDR	MeanL	SDL	MeanU	SDU	MeanB	SDB	MeanG	SDG	MeanCs-137	SDCs-137	MeanX	SDX	MeanZ	SDZ	n
Cultivated Topsoil	W1	8.7	1.5	89.9	15.7	59.0	5.4	20.6	2.8	46.0	8.4	69.6	12.4	6.2	3.8	33.5	5.8	5.6	0.9	44
Cultivated Topsoil	W2	8.6	1.5	87.9	14.9	58.3	5.2	20.6	2.9	44.5	7.4	66.4	12.2	6.1	3.9	32.0	5.8	5.3	0.9	31
Cultivated Topsoil	W3	8.5	1.5	87.1	15.0	58.0	5.3	20.6	3.0	43.9	7.7	65.5	12.2	6.5	3.6	31.6	5.8	5.3	0.9	24
Cultivated Topsoil	W4	8.5	1.5	87.1	15.0	58.0	5.3	20.6	3.0	43.9	7.7	65.5	12.2	6.5	3.6	31.6	5.8	5.3	0.9	24
Cultivated Topsoil	W8	8.3	1.7	82.9	16.1	56.5	5.8	20.4	3.4	41.3	7.4	61.8	12.8	6.4	3.9	29.9	6.1	4.9	0.8	16
Forest Topsoil	W1	6.8	1.6	82.7	27.4	56.2	10.5	17.5	2.8	46.7	19.2	63.4	24.5	14.5	8.5	30.3	11.2	5.8	2.4	13
Forest Topsoil	W2	6.8	1.6	82.7	27.4	56.2	10.5	17.5	2.8	46.7	19.2	63.4	24.5	14.5	8.5	30.3	11.2	5.8	2.4	13
Forest Topsoil	W3	6.8	1.6	82.7	27.4	56.2	10.5	17.5	2.8	46.7	19.2	63.4	24.5	14.5	8.5	30.3	11.2	5.8	2.4	13
Forest Topsoil	W4	6.8	1.6	82.7	27.4	56.2	10.5	17.5	2.8	46.7	19.2	63.4	24.5	14.5	8.5	30.3	11.2	5.8	2.4	13
Forest Topsoil	W8	6.8	1.6	82.7	27.4	56.2	10.5	17.5	2.8	46.7	19.2	63.4	24.5	14.5	8.5	30.3	11.2	5.8	2.4	13
Cultivated Streambank	W1	7.6	0.6	76.8	6.6	55.5	2.2	18.3	1.8	40.8	3.4	58.3	5.0	7.2	9.6	27.9	2.4	5.0	0.4	71
Cultivated Streambank	W2	7.4	0.6	74.6	6.0	55.1	2.2	17.6	1.0	40.5	3.4	57.5	5.4	3.3	1.8	27.5	2.5	4.9	0.4	32
Cultivated Streambank	W3	7.6	0.4	76.3	4.9	55.7	1.8	17.8	0.8	41.4	2.9	58.9	4.5	2.9	1.7	28.2	2.1	5.0	0.3	26
Cultivated Streambank	W4	7.6	0.4	76.3	4.9	55.7	1.8	17.8	0.8	41.4	2.9	58.9	4.5	2.9	1.7	28.2	2.1	5.0	0.3	26
Cultivated Streambank	W8	7.8	0.2	77.8	3.1	56.4	1.0	17.8	0.4	42.5	1.7	60.7	2.5	2.2	0.7	28.9	1.2	5.1	0.2	16
Forest Streambank	W1	5.1	0.3	44.1	3.3	41.0	1.1	16.4	1.2	20.6	0.9	29.0	1.7	17.8	9.9	14.5	0.9	2.5	0.1	8
Forest Streambank	W2	5.1	0.3	44.1	3.3	41.0	1.1	16.4	1.2	20.6	0.9	29.0	1.7	17.8	9.9	14.5	0.9	2.5	0.1	8
Forest Streambank	W3	5.1	0.3	44.1	3.3	41.0	1.1	16.4	1.2	20.6	0.9	29.0	1.7	17.8	9.9	14.5	0.9	2.5	0.1	8
Forest Streambank	W4	5.1	0.3	44.1	3.3	41.0	1.1	16.4	1.2	20.6	0.9	29.0	1.7	17.8	9.9	14.5	0.9	2.5	0.1	8
Forest Streambank	W8	5.1	0.3	44.1	3.3	41.0	1.1	16.4	1.2	20.6	0.9	29.0	1.7	17.8	9.9	14.5	0.9	2.5	0.1	8

Table A.14. MixSIAR discriminant spreadsheet for Upstream Source Sampling using colour coefficients V, R, L, U, B, G, X, Z and radionuclide Cs-137

	MeanV	SDV	MeanR	SDR	MeanL	SDL	MeanU	SDU	MeanB	SDB	MeanG	SDG	MeanCs-137	SDCs-137	MeanX	SDX	MeanZ	SDZ
Cultivated Topsoil	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Forest Topsoil	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Cultivated Streambank	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Forest Streambank	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0

Appendix F: MixSIAR spreadsheet design for Reach Source Sampling

Table A.15. MixSIAR mixture spreadsheet for Reach Source Sampling using colour coefficients V, R, L, U, B, and radionuclide Cs-137 divided by categorical covariate "Year"

Year	V	R	L	U	B	Cs-137
1	9.2	99.6	62.7	20.3	22.5	10.5
1	8.5	96.3	62.5	18.0	20.4	9.9
1	8.8	97.5	62.8	18.4	21.0	8.7
1	8.3	87.7	59.3	18.9	20.6	5.8
1	8.0	84.5	58.8	17.6	19.6	6.6
1	7.9	83.8	58.4	17.8	19.5	8.9
1	7.7	80.8	57.5	17.5	19.1	6.1
1	7.5	78.3	56.3	17.6	18.8	13.6
1	7.6	78.9	56.5	17.7	19.0	9.1
1	9.2	100.6	63.0	20.2	22.4	16.3
1	9.1	100.1	63.1	19.8	22.1	6.8
1	7.8	82.8	58.4	17.0	19.1	9.3
1	7.8	82.8	58.1	17.5	19.2	2.2
1	8.2	87.7	59.3	18.5	20.1	22.7
2	7.4	77.4	56.2	17.1	18.3	14.3
2	7.4	76.7	55.8	17.4	18.6	11.4
2	7.3	74.8	55.2	17.1	18.2	8.1
2	7.4	78.0	56.3	17.4	18.5	7.2
2	7.7	79.9	56.9	17.9	19.3	5.4
2	7.4	77.4	56.2	17.2	18.5	10.3
2	8.1	89.4	60.0	18.1	19.7	8.9
2	7.4	77.5	56.1	17.2	18.4	5.3
3	8.6	91.0	60.3	19.2	21.1	4.3
3	8.0	86.0	59.2	17.6	19.5	15.2
3	9.0	97.9	62.7	19.2	21.8	11.3
3	9.0	99.2	63.2	18.9	21.5	4.5
3	7.7	80.9	57.5	17.5	19.1	3.8
3	7.6	77.4	56.1	17.6	19.0	9.7
3	8.0	84.0	58.5	17.9	19.6	10.3
3	8.6	88.8	59.3	20.0	21.6	4.1
3	7.9	82.1	57.5	18.3	19.8	13.0
4	9.2	99.4	62.5	20.6	22.7	9.8
4	8.5	92.8	61.3	18.2	20.6	11.0
4	8.6	92.4	61.1	18.6	20.9	3.9
4	8.9	91.7	60.6	19.7	21.9	5.1
4	7.7	76.5	55.8	18.1	19.6	10.0
4	7.6	77.6	56.2	17.6	19.0	7.3
4	8.1	83.3	57.8	18.6	20.1	8.6
4	8.9	98.2	62.5	19.5	21.6	5.0
4	7.7	79.8	56.9	18.0	19.4	6.0
5	6.9	71.0	53.9	16.6	17.4	10.7
5	6.8	70.2	53.6	16.5	17.3	4.9
5	8.8	96.0	62.2	19.1	21.4	8.7
5	8.6	95.2	62.2	18.3	20.6	7.4
5	9.4	103.6	64.0	20.4	22.8	7.5
5	9.2	101.2	63.4	19.9	22.3	13.7
5	9.1	101.1	64.0	18.9	21.7	9.6
5	9.5	100.4	62.7	21.2	23.4	0.1
6	8.9	92.5	60.8	19.8	21.9	13.3
6	8.9	92.2	60.7	19.7	21.9	6.8
6	7.8	80.0	57.2	17.5	19.2	7.1
6	7.5	76.3	56.0	17.2	18.8	3.5
6	9.0	98.3	62.6	19.5	21.8	5.4
6	8.4	89.1	59.7	19.1	20.8	5.1
7	6.5	64.8	51.6	16.0	16.6	8.1

Table A.16. MixSIAR source spreadsheet for Reach Source Sampling using colour coefficients V, R, L, U, B, and radionuclide Cs-137

Source	MeanV	SDV	MeanR	SDR	MeanL	SDL	MeanU	SDU	MeanB	SDB	MeanCs-137	SDCs-137	n
Cultivated Topsoil	8.9	1.5	94.8	17.1	60.7	5.9	20.5	2.5	22.2	3.5	6.5	3.6	13
Cultivated Streambank	7.8	0.7	78.8	6.5	55.8	2.1	19.0	2.1	19.9	2.0	10.8	12.2	37
Upstream 2	7.8	1.0	82.3	11.2	57.6	3.9	17.7	2.1	19.3	2.3	8.0	4.6	16
Upstream 8	8.0	1.0	86.7	12.5	59.5	4.9	17.2	1.8	19.4	2.1	4.7	2.7	18

Table A.17. MixSIAR source spreadsheet for Reach Source Sampling using colour coefficients V, R, L, U, B, and radionuclide Cs-137

Source	MeanV	SDV	MeanR	SDR	MeanL	SDL	MeanU	SDU	MeanB	SDB	MeanQ	SDQ	n
Cultivated Topsoil	0	0	0	0	0	0	0	0	0	0	0	0	13
Cultivated Streambank	0	0	0	0	0	0	0	0	0	0	0	0	37
Upstream 2	0	0	0	0	0	0	0	0	0	0	0	0	16
Upstream 8	0	0	0	0	0	0	0	0	0	0	0	0	18

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